

(19) World Intellectual Property Organization
International Bureau



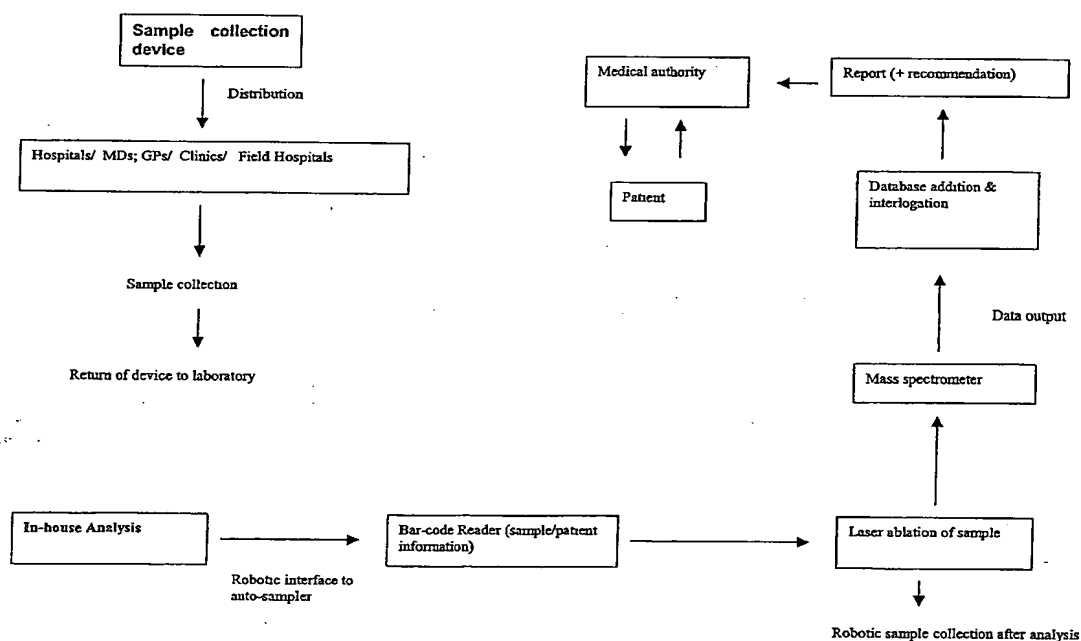
(43) International Publication Date
30 October 2003 (30.10.2003)

PCT

(10) International Publication Number
WO 03/089908 A1

- (51) International Patent Classification⁷: G01N 1/10, 30/72, 33/487
- (21) International Application Number: PCT/AU03/00450
- (22) International Filing Date: 16 April 2003 (16.04.2003)
- (25) Filing Language: English
- (26) Publication Language: English
- (30) Priority Data:
PS 1772 16 April 2002 (16.04.2002) AU
- (71) Applicant (for all designated States except US): DI-
AKYNE PTY LTD [AU/AU]; C/-PH Carey & Co, Level
17, 109 Pitt Street, Sydney, New South Wales 2000 (AU).
- (72) Inventors; and
- (75) Inventors/Applicants (for US only): WATLING, Roger,
John [AU/AU]; 6 August Court, Bullcreek, Perth, Western
Australia 6155 (AU). HERBERT, Hugh, Keith [AU/AU];
4 Niddrie Drive, Toowoomba, Queensland 4350 (AU).
- (74) Agent: BALDWIN SHELSTON WATERS; 60 Margaret
Street, Sydney, NSW 2000 (AU).
- (81) Designated States (national): AE, AG, AL, AM, AT, AU,
AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU,
CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH,
GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC,
LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW,
MX, MZ, NI, NO, NZ, OM, PH, PL, PT, RO, RU, SC, SD,
SE, SG, SK, SL, TJ, TM, TN, TR, TT, TZ, UA, UG, US,
UZ, VC, VN, YU, ZA, ZM, ZW.
- (84) Designated States (regional): ARIPO patent (GH, GM,
KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW),
Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM),
European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE,
ES, FI, FR, GB, GR, HU, IE, IT, LU, MC, NL, PT, RO,
SE, SI, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM,
GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).
- Declaration under Rule 4.17:**
— of inventorship (Rule 4.17(iv)) for US only
- Published:**
— with international search report
- For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: SAMPLE COLLECTING DEVICE AND MASS SPECTROMETRY OF DEVICE



(57) Abstract: A sample collection device comprising a support bearing an inert absorbing matrix for a fluid sample is described. The device may or may not have a lancet. Also described for a sample device is a method of using a mass spectrometer in a laboratory where the sample in its matrix is ionised and the plurality of elements is detected. The results may or may not be quantised in relation to the original sample and an internal ionised reference sample may also be used.

BEST AVAILABLE COPY

WO 03/089908 A1

4 PARTS

SAMPLE COLLECTING DEVICE AND MASS
SPECTROMETRY OF DEVICE

Technical Field

The present invention is concerned with methods and devices for sample collection and simultaneous detection and/or quantitation of multiple trace elements in fluid samples.

Background Art

A wide range of trace metals and other elements is necessary for good health and physical well being in humans and other animals; deficiencies in essential elements have been shown to cause general malaise and lead to the induction of specific disease, commonly resulting in death. For many essential trace elements, it is not simply the absolute concentration, but also the inter-element balances that have a profound effect on health. For example, selenium deficiency is implicated in the aetiology of Iodine Deficiency Disorders amongst humans, whilst copper deficiency, associated with high levels of manganese, may be implicated as a predisposing or causative factor in induction of Bovine Spongiform Encephalopathy (BSE) in cattle and, by association, New Variant Creutzfeldt-Jakob Disease (nvCJD) in humans.

Dietary forages, vegetables, grains and fruits, which fix available trace elements as metal colloids within their tissue, have long been regarded as sources of essential trace elements. Such plant-based metal colloids are about ninety-eight percent absorbed and communities and animals that have a balanced range of plant products as essential components of diet may reasonably be expected to display markedly reduced incidence of specific trace element deficiency-related disease when compared with other groups lacking quality forage or a regular vegetable, fruit and grain intake.

The trace element content of vegetative material is directly related to the bioavailability of essential nutrients in soils supporting the vegetation. Soils vary in their trace element content from enriched to impoverished, according to local geology, soil degradation and nutrient impoverishment and as a function of inappropriate cropping practice, which is widespread throughout the world. In addition, soils throughout the world are sustaining increasing anthropogenic chemical damage threatening the existence of many plants and animals. Consequently, human health is being threatened through the food chain.

While the productivity of the soils may be maintained through the application of N-P-K fertilisers, food crops growing on these soils becomes, without the regular application of biologically-available 'balanced' trace elements, progressively impoverished in essential trace elements and minerals. If not corrected, this may result in sharply increased incidences of mineral deficiency-related disease.

Elements may be classified as being essential or toxic to human and animal health. In the case of animals, trace metal deficiency and/or toxicity is due largely to concentration levels controlled by environmental factors, whereas for humans, both environmental and occupational factors may be important; toxic response may a function of both natural and/or anthropogenic influences.

Ignoring carbon, hydrogen and oxygen, the biologically essential major elements are calcium, chlorine, magnesium, phosphorous, potassium, sodium, nitrogen and sulphur. Essential trace elements include bromine, chromium, cobalt, copper, fluorine, iodine, iron, manganese, molybdenum, selenium, silicon and zinc. If bio-available, many of these essential trace elements induce toxic responses, at elevated levels, or if out of balance with synergistic and/or antagonistic elements. Several other elements (lithium, scandium, rubidium, lanthanum) are minor essential elements.

In addition to dietary trace metal deficiency-induced disease, other cohorts of individuals are occupationally or environmentally exposed to a range of toxic element pollutants, which similarly induce general malaise and/or specific clinical symptoms commonly resulting in complications and death. Notable amongst these are arsenic, lead and mercury, which constitute the top three most hazardous substances on the US Environmental Protection Agency's Toxic Substances and Disease Registry priority list.

The leaching of heavy metals into the aquatic environment, and uptake by wildlife in the food chain, may have a profound impact on human health. Cadmium and mercury, in particular, are strongly bio-accumulated in fish and shellfish.

Although it is not possible to quantify the hazards and deleterious effects associated with all trace elements, some elements clearly present a more serious problem than others. Respectively ranked 1, 2, 3 and 7 on the NPL, arsenic, lead, mercury and cadmium, as elemental pollutants, are considered extremely toxic and the health effects of these elements have received a great deal of attention from research workers. Other elements on the list, in alphabetical order, are aluminium, antimony, barium, beryllium, chromium, cobalt, copper, manganese, nickel, plutonium, radium, selenium, silver, thallium, thorium, tin, uranium, vanadium and zinc.

Unlike many essential trace elements, the concept of a therapeutic index cannot be applied to toxic elements such as lead, cadmium, mercury and arsenic. These toxic elements play no known role in metabolism, as no enzyme has been identified which specifically requires any of them as cofactors. They are extremely hazardous to life and, resulting from ingestion, have been involved in historic poisoning episodes of both human and animal populations. They are increasing in concentration in both aquatic

and terrestrial environments due to anthropogenic inputs, and thus will continue to be a concern to toxicologists and clinicians.

Hence, proactive intervention to identify trace metal and element aberrations within general populations, thereby enabling the early implementation of targeted remedial strategies with consequent minimization of the huge social impact of trace metal-induced disease, is essential. However, mass screening of general populations for trace metal deficiencies and/or toxic metal excesses, with reference to age, sex, socio-economic status and physical geography, while acknowledged as being highly desirable in terms of preventative medicine, is presently impractical. So too, is the mass screening of human food chain components, such as slaughter animals, prior to their entering the food chain.

Present test methodologies require relatively large volumes of fluid samples (for example, 5-10 ml of blood) and are commonly trace element specific, that is, simultaneous measurement of other trace elements potentially present is not possible. Because of this, other relevant trace metals are either overlooked or require further fluid samples for their determination. In the case of blood, this involves invasive, often traumatic extraction, particularly for young children, babies and the elderly, using hypodermic syringes. The derivative body fluid products require stabilisation and preservation, and having regard for transmissible disease such as HIV, appropriate biohazard handling and disposal. Further, the large volumes required give rise to handling and storage problems.

There is no current technology available that can conveniently be used for the collection and broad-spectrum analysis of the trace element content of large numbers of blood and other body fluid samples. Presently available testing methods are cumbersome and expensive, placing the service outside the reach of the general population, particularly in underdeveloped regions where problems are often greatest. Further, there are no convenient and sensitive mass spectrometric methods for detecting pollutants or contaminants in fluids such as water or lubricants.

There is therefore a need for improved methodologies which will enable more efficient and cost effective screening of trace elements in fluid samples.

It is an object of the present invention to alleviate at least some of the disadvantages of prior art methods, or to provide a useful alternative.

Summary of the Invention

According to a first aspect there is provided a sample collection device comprising an inert collection matrix capable of adsorbing or absorbing a fluid sample, and a solid support, wherein the inert matrix is affixed to an area of the solid support

Particularly useful matrices may be selected from aragonite, aluminium hydroxide, titania, glucose, Starch "A", Starch "B", glucodin, cellulose powder/granules, fibrous cellulose, hydroxy butyl methyl cellulose, vegetable flour and the like, or mixtures thereof. Particularly preferred is fibrous cellulose. The fibrous cellulose matrix
5 may be modified by oxidation and/or acid hydrolysis to improve its properties and thus provide enhanced reproducibility and sensitivity.

The vegetable flour may be selected from rice, maize, wheat, soy, rye or corn flour, or mixtures thereof. Particularly preferred is rice flour.

The inert matrix may also contain, on or within, one or more pre-calibrated
10 selected analytes as internal standard, to aid in the quantitation of trace elements in the sample applied to the collection device.

The device of the present invention may also comprise an integral lancing member, capable of piercing for example skin or tissue, to aid in the collection and application of a blood or body fluid sample to the inert matrix. The lancing member may
15 be mounted adjacent to, within or below the area of inert matrix. There may be included a guiding channel in the inert matrix, to guide the lance should it be disposed below the inert matrix area.

The device may also be equipped with a laser-scannable bar code which may contain patient information or other information concerning the sample, its nature and
20 source. The device may also include an antibiotic barrier, to prevent contamination of the sample to analytical equipment and personnel.

Preferably the inert matrix is applied to only one side of the support. It is also preferred that the area to which the matrix is applied is smaller than the area of the solid support and that it be in the shape of a small tablet-sized disc.

25 The inert matrix may include hydrophobic and/or hydrophilic components, depending on the nature of the sample and the analysis to be performed.

Preferably the solid support is made of flexible material having sufficient durability to withstand transport and handling. Of course it will be understood that the support can be made of rigid material, depending on the nature of application. It is also
30 preferred that the device is of sufficiently small size to allow transport of the device through mail and for ease of storage. The device may have an integral or separate cover sheath, to protect the inert matrix and prevent possible contamination after collection. The cover sheath also protects the device during transport and handling.

According to a second aspect there is provided a sample collection device having
35 multi-layer construction wherein the collection matrix layer is sandwiched between two

supporting layers, one of said supporting layers having an opening, which exposes an area of the collection matrix.

Alternatively, the sample collection device may encapsulate a collection matrix tablet within the body of the support wherein the matrix is exposed flush with one
5 surface of the support.

The collection device and methods of the present invention may be used for analysis of any fluid sample, including body fluids, oils and other lubricants, water from drinking supplies as well as waste water, and the like. Body fluids such as whole blood are particularly preferred, however, separated blood (eg. plasma or serum) and other
10 body fluids, such as urine or sweat, can also be used with the same device.

It will be understood that a sample of body fluid, particularly blood, can be collected for analysis by conventional means, or by using for example a sample collection kit comprising a resealable, sterile sample collection device, embodying a bar coded support in which is embedded, or to which is affixed, a tablet, wafer, wad, strip or
15 the like, of sample absorption/adsorption matrix, a sealed alcohol-saturated wipe, and a separate retractable, single use, spring-loaded lance for penetrating the skin and drawing blood. Of course a lance can be omitted from the kit if the sample to be collected is for example urine or sweat.

As indicated above, the analytical sample need not be a body fluid. Thus, the
20 devices and methods of the present invention are equally applicable to collection and analysis of water or oil samples without significant adaptation of collection devices or analytical procedures and equipment.

The matrix of the sample collection device can include one or more matrix-matched standards either adsorbed/absorbed onto/into sample collection matrix or,
25 alternatively, supported on an impermeable substrate. Here, the matrix may be spiked with elements, for example, Be, In and Hf and these elements will serve as internal standards that will be released simultaneously with the sample during ablation; this will facilitate matrix matching.

According to a third aspect there is provided a method of detecting
30 simultaneously a plurality of elements in a fluid sample adsorbed onto or into an inert collection matrix, comprising:

(i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample, and

(ii) detecting plurality of elements in the ionised portion of the sample by mass
35 spectrometry.

According to a fourth aspect there is provided a method of quantifying simultaneously a plurality of elements in a fluid sample adsorbed onto or into an inert collection matrix, comprising:

(i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample;

(ii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;

(iii) measuring quantity of ionised portion of sample, and

(iv) determining quantity of the plurality of elements in the sample.

According to a fifth aspect there is provided a method of quantifying simultaneously a plurality of elements in a fluid sample adsorbed onto or into an inert collection matrix having an internal standard applied thereto, comprising:

(i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample and a portion of said internal standard;

(ii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;

(iii) measuring quantity of ionised internal standard in the ionised portion of the sample by mass spectrometry, and

(iv) determining quantity of the plurality of elements in the sample with reference to quantity of ionised internal standard.

According to a sixth aspect there is provided a method of quantifying simultaneously a plurality of elements in a fluid sample adsorbed onto an inert collection matrix, comprising:

(i) introducing into the fluid sample a known quantity of a measurable internal standard

(ii) exposing the sample to high energy radiation capable of ionising at least a portion of the sample and the internal standard;

(iii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;

(iv) measuring quantity of ionised internal standard in the ionised portion of the sample by mass spectrometry, and

(v) determining quantity of the plurality of elements in the sample with reference to quantity of ionised internal standard.

According to a seventh aspect there is provided a method of quantifying simultaneously a plurality of elements in a fluid sample adsorbed/absorbed onto or into an inert collection matrix comprising:

(i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample;

(ii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;

5 (iii) exposing a matrix-matched Certified Reference Material (CRM) to high energy radiation capable of ionising at least a portion of the CRM;

(iv) measuring quantity of ionised CRM in the ionised portion of the sample by mass spectrometry, and

10 (v) determining quantity of the plurality of elements in the sample with reference to the CRM.

According to an eighth aspect there is provided a method of quantifying simultaneously a plurality of elements in a fluid sample supported on an impermeable substrate, comprising:

15 (i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample;

(ii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;

(iii) exposing a matrix-matched Certified Reference Material (CRM) to high energy radiation capable of ionising at least a portion of the CRM;

20 (iv) measuring quantity of ionised CRM in the ionised portion of the sample by mass spectrometry, and

(v) determining quantity of the plurality of elements in the sample with reference to the CRM.

25 Details of some useful CRM's, for example, SARM 1, 3 and 46 (South African Bureau of Standards), and SY-2 (Canadian Certified Reference Material Project (CCRMP)) are given in Table 1. Other standard element cocktails may include elements such as Be, In, Hf, Bi, Th to cover the mass calibration range, but may include any element as a standard, that is not being analysed.

30 Preferably, the sample is whole blood and sample size is approximately 50 μ l to 100 μ l and even more preferred size of sample is 50 μ l or less. Of course, separated blood may also be used, eg. plasma or serum.

Also preferred is that the high energy radiation is UV laser radiation and that the sample is exposed to such radiation for a period of approximately 30 seconds, but may be between 10 and 120 seconds. The devices and methods of the present invention
35 may be used in conjunction with any Inductively Coupled Plasma-Mass Spectrometer

(ICP-MS) system. Particularly preferred are quadrupole and Time-of-Flight (TOF) ICP-MS systems.

The preferred elements to be detected and/or quantified are dietary trace elements, toxic elements and markers of pollution or wear and tear. For blood and other body fluids, these elements can include Li, Na, Mg, Al, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Cd, Sn, Sb, Te, Ba, La, Ce, Eu, Dy, Yb, Hg, Tl, Pb, Th and U. For wear metals in lubricants such as oil, the element array may include Li, B, Mg, Al, Si, P, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Sr, Y, Zr, Mo, Ag, Cd, Sn, Sb, Ba, La, Ce, Hf, Hg, Pb, and U.

In a preferred embodiment the matrix or the support comprise one or more wells or indentations to accommodate the fluid sample.

According to a ninth aspect there is provided a method of collecting a fluid sample for mass spectrometry analysis of multiple element content comprising the application of the sample to an inert matrix having a low background element content, wherein the matrix is selected from the group consisting of aragonite, aluminium hydroxide, titania, glucose, Starch "A", Starch "B", glucodln, cellulose powder/granules, fibrous cellulose, hydroxy butyl methyl cellulose, vegetable flour or mixtures thereof.

Description of the Preferred Embodiment

The present invention is in part based on Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry technique, which allows rapid, automated, cost effective mass screening of general populations, bloodstock, zoo animals, pets and slaughter animals to identify trace element aberrations in body fluids. This technology facilitates proactive remedial intervention to target and correct essential trace element imbalances and/or toxic heavy metal excesses and enables identification and rejection of heavy metal-contaminated slaughter animals designed for human consumption. The methods and devices of the present invention are also useful for detection and quantitation of trace elements, metals and the like in fluids such water and lubricants, as indicators of for example water pollution or mechanical wear and tear.

The present invention in its various embodiments allows the simultaneous analysis and/or quantitation of a broad spectrum of up to 50 trace elements during a primary analytical run. A secondary run, using a screened torch may include Ca, Mg, Na, K and Fe. The analytical cost of a sample is lower than that of a large number of single element analyses currently being performed, on a chemically unmodified 50-100 micro-litre volume of body fluid sample or other fluid sample (single drop) adsorbed onto an inert collection matrix. In case of blood, the sample collection device, and collection protocol, may be so configured to eliminate the use of hypodermic syringes, and hence

potential for stick injuries, is non-invasive and hence, non-traumatic, and does not involve the preservation, movement and storage of large volumes of blood and urine, or involve large biohazard disposal facilities. Indeed, in the case of humans, samples may generally be self-acquired at any geographic location through absorption/adsorption of a drop of biological fluid, such as blood from a pin prick, into/onto a lightweight collection device as described herein, and dispatched to the nearest analytical facility by post or courier. Because an approximately 8000°C argon plasma is involved in ionisation of the samples, the body fluid samples are expected to be largely sterilized during analysis.

Certain embodiments of the present invention have been developed using an ultraviolet laser and quadrupole inductively coupled plasma-mass spectrometer (LA-ICP-MS) with manual sample handling. However, the present methods are equally applicable to Time-of-Flight (ToF) and High Resolution mass spectrometry techniques. Further, the methods of the present invention, whether they make use of quadrupole, ToF or High Resolution mass spectrometry, can be automated to allow rapid, high volume throughput screening of samples.

The methods and devices of the present invention permit cost effective, simultaneous, automated mass screening of blood, and other body fluids, for a wide range of essential and toxic trace elements on micro-litre volumes of test fluid absorbed onto inert collection matrices. In certain preferred embodiments the core of the analytical system comprises a quadrupole Laser Ablation-Inductively Coupled Plasma-Mass Spectrometer. The spectrometer may be used in conjunction with an associated automated sample insertion system.

In preferred embodiments of the present invention the collection device, or kit of parts, is envisaged to consist of the following components:

- housing mount that forms the surround of the actual collection matrix and acts as the support of this matrix and also increases robustness of the entire device allowing for transport of the entire system;
- the collection matrix itself consisting of an absorptive pellet;
- a mechanism for puncturing skin and facilitating the collection of a single drop of blood; and
- a bar code or equivalent which ultimately will facilitate the recognition of both the sample and its association with the client.

However, the collection device, or kits of parts, may exclude certain features or include additional features.

The invention will now be described in more detail with reference to non-limiting examples.

Examples

Example 1: Sample collection and application

5 Samples may be collected and applied to a chosen collection matrix of the present invention in a conventional manner well known in the art.

For example, blood from a subject may be collected using a kit which comprises a shielded, retractable, spring loaded 'pricker', as part of the sample kit, which also includes a sealed, alcohol-saturated wipe, or swab, for pre-cleaning the skin area to be
10 pricked to avoid unnecessary sample contamination.

It will be understood however that collection of samples of other body fluids, such as urine and sweat, or other fluids such as water or oil and other lubricants, will not require most of the components stipulated above for blood collection, but it will nevertheless be important to exclude contaminants. Conventional techniques for this
15 will be known to those skilled in the art.

The fluid sample, whichever fluid may be of interest, can be applied to the collection matrix for analysis by any known means. For example, a particular quantity may be applied to the collection matrix by a pipette, a capillary tube, a dip-stick or similar device. Exact quantity applied is not important but may be controlled if desired.

20 Alternatively, particularly for blood sample collection, a collection device such as described in Example 2 below may be used.

Example 2: Sample Collection Device

An example of one type of sample collection device of the present invention, particularly suitable for collection of a blood sample, incorporates an inert fluid
25 absorption matrix, most preferably a fibrous cellulose matrix (Whatman 540, but also 541, 542 and other cellulose filter papers, Whatman International Ltd, Maldstone, England), typically shaped in the form of a small tablet-size disc. The matrix is affixed to or encased within a small, lightweight, disposable or re-cyclable holder (disc holder or solid support material). Ideally the holder is made of relatively rigid material (for example
30 plastic, cardboard or similar material). The device is designed so that a drop of blood or body fluid can be placed on the absorption matrix and the device sealed at the site of collection. Thus immobilized sample can be easily transported via post or courier to a sample analysis center and/or stored.

Of course the device may be used for other samples, which are not body fluids.
35 For example water or a lubricants.

A collection device of this embodiment of the present invention, incorporating a number of features described below, is depicted in Figure 1. In plan view (A) the device is typically rectangular in shape and has an area of absorbent collection matrix (1) disposed on the surface, and may also have a bar code (2) containing relevant information about the sample and/or the subject. The collection matrix is preferably fibrous cellulose but other matrices described hereafter may also be used. The collection area shown is circular in shape but may be any other suitable shape. A cover sheath (B) may be provided, to cover the collecting matrix area after the sample has been collected. Figures 2 and 3 show the collection device in cross section, in closed and open positions respectively. The carrier or backing (support) portion (A) of the device can be suitably made of plastic or some form of card (stiff paper, cardboard and the like) material. The cover sheath (B) may be made of similar materials. Both the backing portion and the cover sheath may include a locking ridge (3), for positive engagement between the backing and cover sheath, and also to prevent the cover sheath, if used, from sliding off entirely.

Figures 2 and 3 also show the area of collection matrix (1) and a stylus or lance (5) disposed below the collection matrix and within the carrier or backing material. The lance may be guided by a channel (4) in the collection matrix, so that when the device is pressed between the thumb and a finger, the lance will be forced through the channel and into the finger, thus piercing the finger and enabling a sample of blood to be collected onto the collecting matrix. Once the sample has been taken, the cover or sheath can be slid over the collecting matrix, thus protecting the sample as well as individuals handling the used device.

Figure 4 is an enlargement of a section of figures 2 and 3, showing in more detail the preferred arrangement of the lance, collection matrix and the guiding channel.

Typically, a collection device contemplated herein, in a particular preferred configuration, will have dimensions of approximately 40x20 mm and will be about 2 mm thick. However, larger or smaller collection devices may be useful in different applications and can be designed along equivalent parameters.

The collection device is primarily designed for the collection of blood and other body fluids prior to analysis of the trace element content. However, similar design principles can be used for sample collection of other fluids, omitting the integral lance. Of course, even for blood sample collection, the device described above may be provided with a separate lance, packaged together in a kit of separate components if desired.

The design of the sample collection device provides for low manufacturing costs, a robust configuration, ease of transportation, ease of storage, and can be used to collect a drop of test sample from a remote site by an inexperienced collector.

The matrix, which forms an integral part of the device, is typically an inert material with respect to fluid interaction prior to analysis and does not interfere with the subsequent sample analysis. The sample adsorbed onto or into the matrix can be stored indefinitely, without the addition of preservatives that may add contaminants to the sample.

The preferred material suitable for the matrix is cellulose, either granular or fibrous and may be either formed or preformed. Typically, the sample of blood transferred to the blood collection device does not have a specific volume. Hence the matrix may be encoded with an internal standard to normalize the analytical data on analysis.

The matrix may also be composed of inorganic materials suitable for a matrix of the ceramic-type, for example compounds of lithium, boron, carbon, magnesium, aluminium and silicon. Although this list is not exhaustive, it does encompass the main ingredients for an appropriate robust thermo-ceramic.

Typically, a sample of blood is transferred to the collection device that has a small lance or puncturing needle incorporated into the matrix, or into the backing/support material. The patient grips the device and causes a small pinprick to be administered. The collected blood does not have to have a specific volume as the matrix can be encoded with an internal standard, which normalizes the analytical data on analysis.

The device can have a laser-scannable bar code for recognition of the patient or to include any other additional information on the sample and its source. The amount of blood required is usually less than 50 μ L. The device can also have a sealing mechanism to ensure that the device plus sample can be transported and will not be contaminated.

The matrix may be affixed to, or encapsulated within, the support material or holder by any known means and may employ adhesives. Further, an antibiotic barrier may be applied to prevent contamination of the sample or the analytical equipment and personnel.

The present invention also makes use of collection devices which do not possess a collection matrix affixed thereto. The collection matrix may be simply omitted and the sample applied directly to the support material (backing). This may be particularly useful in certain body fluid collection devices. In such devices it may be advantageous to

introduce indentations (wells) into the support material, to allow for sample immobilization or the application of multiple samples and/or standards to the same support material (device) by application to multiple indentations (wells) in the support material.

5 Sample of fluids applied to any of the collection devices describe herein may be dried before analysis.

Example 3: Sample Analysis System

Traditionally, quantitation in LA-ICP-MS has been approached by controlling the power coupling the laser to the sample, to ensure uniform ablation characteristics and transfer of uniform amounts of solid to the analytical plasma. While this has much to
10 recommend it when the nature of the matrix can be assured (eg. glass or similar), there are significant problems associated with standardisation of the coupling and transfer efficiency when matrices are not uniform. Furthermore, when the surface characteristics of the sample also vary it is extremely difficult to ensure uniform ablation.

15 Until the present invention laser ablation ICP-MS technology has been at best a semi-quantitative technique and more usually a comparative technique for the determination of trace element levels in any solid material. In this embodiment of the invention quantitation in LA-ICP-MS has been approached by quantitation of the amount of debris (ablated or ionised material) that is actually transported from the laser cell to
20 the analytical plasma.

When using an Infrared laser, where the particle size of ablated material is relatively large, Ultra-violet spectral interference can be used to quantify the amount of particles (ablation efficiency) entering the plasma. However, in the majority of cases the techniques currently employ either UV or Excimer lasers. These lasers produce
25 particles that are too small to have sensible UV scattering and consequently relatively inexpensive particle quantitation is not possible. However, laser interferometry can be used, as an appropriate alternative technique, to quantitate the amount of ablated material and thus the efficiency of UV lasers. Once transport efficiency is quantified, it is then possible to quantify the amount of particles that are entering the analytical plasma and hence quantify the resulting signal (ie. amount of any one element).

30 The quantification process can be further enhanced by using internal standards in the support matrix of the collection/transportation device described above, or by adding one or more standards to the sample to be analysed. A suitable internal standard can be selected from elements which are not commonly present or are below detectable levels in a particular sample. Thus, for blood samples, elements such as Hf, Ir, Ru, Rh, Ta and heavy rare earths can be used as internal standards, and

incorporated into the inert matrix by bonding to the surface of the particles used to produce the matrix, or may even be present as a natural constituent of the sample itself.

In case where the internal standard is incorporated into the matrix, when the sample is ablated, the particles of the matrix are carried into the analytical plasma along with the sample. Quantitation of the transport efficiency of all debris is achieved using
5 laser interferometry, or an appropriate alternative technique, and supported by normalisation to the signal from internal standards. Since the bonding characteristics of the internal standards and the efficiency of absorption of the matrix are known, as is the transport efficiency, it is possible to calculate the concentration of the element in the
10 sample adsorbed onto the matrix, in this case blood.

In another embodiment of the present invention, quantitation by LA-ICP-MS has been approached by quantitation against matrix-matched standards.

Quantitation is achieved by using internal standards in the collection matrix, or by adding one or more standards to the sample to be analysed. A suitable internal
15 standard can be selected from elements that are not commonly present or are below detectable levels in a particular sample. Thus, for blood samples, internal standards are incorporated into the inert matrix through solution doping, or may even be present as a natural constituent of the matrix itself. The collection matrix is doped with the relevant standards to act as mass calibration standards. These may be Be, In and Bi, or
20 other suitable combination depending upon the analysis required. In addition any other analyte can be spiked into the matrix pad and the pads analyzed. The spiking of calibration standards onto the matrix pad allows for its analysis as a "blank". To the standard-spiked matrix pads, blood, sweat, urine or any other fluid sample may subsequently be added. The sample is dried at 105°C for 2 hours, but may be any other
25 suitable temperature and time, and then ablated. The sample plus the 'under' matrix is ablated and carried into the plasma simultaneously. Ionization is achieved for both components and, in this way samples are calibrated. Hence, because of this, the nature of the sample is not important as the sample and the matrix containing the internal standards are introduced simultaneously to the plasma. This protocol removes the
30 necessity for a spike as the spike is already in the matrix pad on which the sample is collected. Therefore, it does not matter what the sample is, as it will be introduced into the plasma with the standards thereby overcoming any matrix interference. In this embodiment, it is not necessary to add a range of analytes to the matrix because the Be, In and Bi act as the calibrants and can be calibrated against all other elements with
35 respect to mass response before the samples are analyzed. Of course there are a series of matrices that are spiked (detailed in text already) with standards from which

calibration curves may be established thereby facilitating quantification of trace elements contained in the blood or other fluid.

Thus, fibrous cellulose matrix pads are prepared and doped with the set of mass calibration elements and dried. Blood, or other fluid is added, dried and ablated using a 10x10 matrix raster. The data are collected and read against results obtained from a concentration range (100, 200, 500ppb etc) of multi-element standards prepared and measured in the same way. Quantitation for any matrix may thus be achieved because the standard and sample are being introduced in the same way which therefore negates potential matrix problems. The data are cross-referenced to Be, In and Bi in the standards and in the matrix with sample, and their relative values in each normalized.

The core components of the Sample Analysis System of this embodiment comprise a laser for producing an aerosol of the sample (Laser Ablation), an argon plasma, or 'electrical flame', operating at temperatures in excess of 7,000°C (Inductively Coupled Plasma) in which the aerosol is ionized, a mass filter (Mass Spectrometer) for separating the ions into 'packets' according to their mass to charge ratio, and an ion detector (Multi-channel Analyzer or Ion Multiplier) for detecting the ions in each 'packet'. The system operates with a routine sensitivity capable of achieving parts per billion detection limits. All data can be electronically stored for future reference.

Suitable ICP-MS system utilizes a quadrupole mass filter, controlled by alternating RF and DC fields in the quadrupole, to allow transmission of ions of one selected mass to charge ratio at any specific time. Cycling of the quadrupole allows passage of any selected ion with a mass to charge ratio of <250amu at specific times during the cycling program. Each naturally occurring element has a unique and simple pattern of nearly integer mass to charge ratio, corresponding to its stable isotopes, thereby facilitating identification of the elemental composition of the sample being analyzed. The number of registered element ions from a specific sample is proportional to the concentration of the element isotope in the sample.

For multi-element analysis, the quadrupole is generally configured to scan at 1Hz (once per second). Under this circumstance, if, for example, 100 isotopic masses are being analyzed, each isotopic mass will be collected only one hundredth of the entire scan time.

It will be understood that other configurations and types of instrumentation can be used with the devices and methods of the present invention without undue modification of protocols presented herein.

In one exemplary operation, the sample is introduced into a laser ablation cell and ablated, using either an Excimer or Frequency Quadrupled Nd-YAG laser, for a

period typically not exceeding 30 seconds. Debris from the ablated sample passes down an interface tube, made from Nalgene as a suitable plastic material but other material could also be used, attached to the torch of an inductively coupled plasma (ICP). The sample debris passes through a zone in this tube, adjacent to the torch, into
5 which independent laser radiation is being passed. A concentric series of dynode detectors measures the photon flux, reflected from the sample debris particles, which facilitates quantitation of particle scattering. Knowing the amount of scattering allows linear correlation to the amount of particles doing the scattering. The Laser scattering device is calibrated using conventional smoke cells.

10 The level of scattering is a quantitative indication of the amount of debris passing down the tube. This debris contains the sample material (blood) in addition to particles of a pre-coded (with internal standard) carrier matrix. The particles now pass on into the Inductively Coupled Plasma (ICP) where they are ionised and separated using Time of Flight (ToF) segregation. The elemental composition for the sample is
15 established and quantified with reference to the signal obtained from each of the analyte isotopes. Quantitation of the concentration of elements present in the sample and hence the blood, is calculated with reference to the scattering signal from the Laser Interferometer. The amount of sample being analysed is normalized to the signal generation by ionisation of the components in the pre-coded matrix. In this way the
20 amount of material ablated is used to obtain the mass component of the transported material and the elemental signature of the pre-coded matrix facilitates normalization of the response with reference to an ionisation efficiency cross comparison.

Quantitation of elements in the sample may also be achieved by incorporating standards into the sample or into/onto the collection matrix/support, or both. The pre-
25 coded collection matrix may contain a cocktail of elements that are not naturally present in the sample such as blood or other fluid, at levels above the detection limit of the technique. These elements typically include one or more (i.e. mixture of) Beryllium, Scandium, Zirconium, Niobium, Rhodium, Ruthenium, Indium, Hafnium, Tantalum, Rhenium, Osmium and Iridium. This requires doping of appropriate analytes at levels
30 between 1 and 10,000 ng/mL to the matrix or support. The elements are chosen to cover both mass and ionisation potential ranges present in the analytically significant analytes.

In another exemplary operation, the sample is introduced into a laser ablation cell and ablated, using a Frequency Quadrupled Nd-YAG laser operating at 266 nm, for
35 a pre-determined time interval typically dictated by the number of analytes being acquired. Debris from the ablated sample passes down an interface tube, made from

Nalgene or suitable other plastic, attached to the torch of an inductively coupled plasma (ICP). The pre-coded matrix may contain a cocktail of elements that are not naturally present in blood, at levels above the detection limit of the technique. These elements typically include one or more (i.e. mixture of) Beryllium, Scandium, Zirconium, Niobium, Rhodium, Ruthenium, Indium, Hafnium, Tantalum, Rhenium, Osmium and Iridium. This requires doping of appropriate analytes at levels between 1 and 10,000 ng/mL to the matrix. The elements are chosen to cover both mass and ionisation potential ranges present in the analytically significant analytes.

Readout from the spectrometer, for reporting purposes, is expressed in concentration units appropriate to clinically accepted protocols. In addition, the readout contains information on the acceptable ranges of analytes in normal healthy individuals and indicate whether the sample under investigation is below, above or in the accepted range.

The methods and devices of the present invention enable the mass screening of a variety of blood or other body fluid samples for a wide range of essential and toxic trace elements, or of samples of other fluids such as water or lubricants, for contaminants indicative of pollution or wear. Only a small volume of sample liquid (one or two drops) is required for multiple element analysis. Sample collection of body fluids does not require the use of a hypodermic needle and consequently is essentially non-invasive and considerably safer than existing methods. The sample is collected and stored in an inert matrix without need for addition of preservatives. The sample can be handled and transported safely and easily. The preferred method of analysis, quadrupole Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry, is very sensitive and can detect and measure trace/ultra trace amounts of an element. The methods described herein are suited to full automation and high throughput screening and analysis of samples. Further, the methods and devices of the present invention enable multi-element testing at a significantly lower cost than many current single element tests, thus making the economical mass-screening of target populations possible.

Examples of suitable internal standards which may be used for quantitation of elements, in conjunction with the devices and methods of the present invention, are detailed in Table 1 below.

Table 1:

Sample Name	SARM 1	SARM 3	SARM 46	SY-2
Alt. Name	NIM-G	NIM-L	S14	
Sample Type	Granite	Lujavrite	Stream Sediment	Syenite Rock

	ppm	ppm	ppm	ppm
Si	353848	244936		280975
Ti		2878		899
Al	63933	72180		63722
Fe 3+	4197	61410		16986
Fe 2+	10105	8784		27672
Mn	155	5963		2478
Mg	362	1889		18222
Ca	5575	23013		58888
Na	24926	62093		31974
K	41424	45741		36942
P	44	262		1877
Ag				0.029
As	19.3	1.92		17.3
Au	0.0011	0.00084		0.00052
B				88
Ba	120	450		460
Be	7.75	29.5		22
Bi	0.275	0.468		0.111
Br				
Cd	0.113	0.91		0.21
Ce	195	240		175
Cl	263	1200		140
Co	0.36	2.44	54	8.8
Cr	12	10	593	9.5
Cs	1.08	2.78		2.4
Cu	12	13	583	5.2
Dy	17	3.1		18
Er	10.5	2.6		12.4
Eu	0.35	1.2		2.42
F	4200	4400		5030
Ga	27	54		29
Gd	14	3.8		17
Ge		0.89		1.3
Hf	12.4	231		7.7
Hg	0.0189	0.0445		0.0043
Ho	3.6	0.9		3.8
I				
In				
Ir	0.0005			0.0005
La	109	250		75
Li	12	48		95
Lu	2	0.4		2.7
Mo	2.84	1.21		0.53
N				
Nb	53	960	26	29
Nd	72	48		73
Ni	8	2.2	122	10
Os				

Pb	40	43	14000	85
Pd	0.007			0.015
Pr	19.5	16.4		18.8
Pt				
Ra				3.7
Rb	325	190	18	217
Re				
Rh				
Ru	0.01			0.002
S		850		160
Sb	1.19	0.13		0.26
Sc	0.9	0.5		7
Se	0.012	0.014		20
Sm	15.8	5		16.1
Sn	3.3	7.4		5.7
Sr	10	4600	28	271
Ta	4.9	25.2		2.01
Tb	3	0.7		2.6
Te	0.007	0.009		0.002
Th	51	66		379
Tl	0.93	0.325		1.5
Tm	2			2.1
U	15	14		284
V	2	81	195	60
W	1.45	8.28		0.76
Y	143	22		128
Yb	14.2	3		17
Zn	50	395	6200	248
Zr	300	11000	95	280

The collection matrix, if one is used, may be impregnated with a trace metal cocktail, of known concentration using purpose prepared aqueous solution standards. In certain preferred embodiments, the matrix may contain 2ppm of Be, In, Hf as internal standards to calibrate the mass response for the system in blood analysis. In other embodiments describing wear metal analysis of oil, 2ppm of Be, In and Th may be used. In yet other embodiments, different suites of elements may be used.

Separate standard matrix pads may be used to calibrate the sensitivity and these may be as follows for blood and body fluids: a single pad containing, but not restricted to, Li, Na, Mg, Al, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Cd, Sn, Sb, Te, Ba, La, Ce, Eu, Dy, Yb, Hg, Tl, Pb, Bi, Th and U at 1 ppb, a second pad with all these at 2 ppb. A third pad with all of these at 5ppb a fourth pad with all of these at 10ppb a fifth pad with all of these at 20 ppb a sixth pad with all of these at 50 ppb a seventh pad with all of these at 100ppb an eighth pad with all of these at 200ppb a ninth pad with all of these at 500 ppb a tenth pad with all of these at 1000ppb. An appropriate

concentration can then be used for the set of elements being determined in a particular fluid sample. In another embodiment, a suite of elements appropriate to wear metal analysis in oil, for example, Li, B, Mg, Al, Si, P, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Sr, Y, Zr, Mo, Ag, Cd, Sn, Sb, Ba, La, Ce, Hf, Hg, Pb and U may be doped into matrix pads at 1ppb through 1000ppb as above. so that when ablated, a range of elements across the mass spectrum may be used as internal standards to standardise the system. Thus, the collection matrix, when used, may contain a pre-calibrated concentration of selected analytes. Both a broad-spectrum general collection matrix/device and a test specific matrices/device/s may be employed for specific elements or suites of elements. Further, any one, or combination or range of internal standards analytes may be spiked into the collection device to ensure its broad spectrum or specific use. For example, for broad spectrum, the preferred combination is , Li, Na, Mg, Al, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Cd, Sn, Sb, Te, Ba, La, Ce, Eu, Dy, Yb, Hg, Tl, Pb, Bi, Th and U and for specific applications, for example analyzing oils preferred is , Li, B, Mg, Al, Si, P, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Sr, Y, Zr, Mo, Ag, Cd, Sn, Sb, Ba, La, Ce, Hf, Hg, Pb and U and for blood the preferred combination is , Li, Na, Mg, Al, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Cd, Sn, Sb, Te, Ba, La, Ce, Eu, Dy, Yb, Hg, Tl, Pb, Bi, Th and U.

A typical procedure of collecting and analyzing a sample is summarized in Figure 5. Of course, manual procedures can also be adopted, as can variations of the proposed exemplary scheme.

Example 4: Analysis of collection matrices

The purpose of the experiments described below was the definition and/or refinement of chemically and mechanically robust fluid adsorption/absorption matrix/matrices to facilitate the collection and quantitative analysis of micro-litre fluid samples by Laser Ablation-Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS). For purposes of this example fluids under consideration are blood, urine and oil. However it will be understood that any other fluid, biological or otherwise, may be analysed using similar matrices and techniques.

Preferably the sample collection matrices should be suitable for incorporation into a robust, transportable sample collection device. The device should have specific attributes such as but not limited to:

- be cheap and capable of precision mass production;
- be small and easily accommodated in laser cells for ablation prior to analysis;

- be able to be coded for automatic pre-analysis reading and referral of the sample back to the data, and the data to the client;
- for blood collection, contain a mechanism for penetration of individual patient's skin thereby minimising potential 'stick injuries'. There would be some form of shielding device, or mechanism, that would "shield" the puncturing mechanism such that it would not be able to penetrate the skin of another person subsequent to initial collection of blood;
- produce minimum biohazard with material after analysis and prior to disposal. This implies a small collection device and a small blood sample (less than 100 μ L), and a very small amount of material comprising the sampling device itself that would ultimately have to be incinerated;
- easy transportability to and from the collection site and through conventional mailing procedures. The device should be such that conventional postal systems can be used without the possibility of contamination and release of potentially bio-hazardous material; and
- be capable of being used by non-medical personnel.

MATRIX MATERIALS

The original preferred matrix material used for process testing was fibrous cellulose. Using this material, it was possible to readily form backed cardboard 'punch-outs' containing the cellulose absorptive medium. Micro-litre samples of blood, added to this material, were qualitatively analysed by LA-ICP-MS. Qualitative spectra and raw count data were generated, much of which reflected trace metals in the absorbed blood. However, it was reasoned that the cellulose, being a natural organic product, might be contributing to the analyte signal of a range of elements recorded. Hence, it was determined that cellulose, together with an array of other potential matrix materials, be further investigated, both in terms of its chemical and physical characteristics.

Some attributes of suitable sample collection matrices include but are not limited to:

- must be chemically "clean", that is, have a low concentration of analytes of interest;
- robust, that is, capable of transportation, often over long distances without fragmentation;
- have significant wettability, both by aqueous and non-aqueous (blood and oil) samples while still retaining integrity;
- capable of withstanding laser ablation removal of samples; and
- not contribute to analyte segregation during analysis.

MATRIX CHOICE

The parameters detailed above govern the choice of matrix and, as such, preclude certain materials. A list of matrices investigated follows with indications as to their potential suitability, or otherwise, which resulted in a final short list of potentially useful material to be subsequently tested. The choice of white metal oxides as potential matrices is based on the fact that the two detailed herein are locally manufactured in bulk, are extremely cheap and, using the modern generation of UV lasers (unlike IR lasers), are customarily considered not to have variable coupling efficiencies between light and dark matrices.

Potential organic and inorganic matrix materials investigated are:

- Pig-toe mussel shell (aragonite) – sourced from the WA pearl industry
- Aluminium hydroxide – Alcoa (WA)
- Titania – New Millennium (WA)
- Bacterial grade glucose – sourced by Professor Watling
- Starch "A" - BDH Analar analytical reagent
- Starch "B" – Ajax Chemicals Univar analytical reagent
- Glucodln – Boots Healthcare Australia
- Cellulose – high purity powder – Sigma Chemicals Microgranular
- Cellulose – high purity fibrous cellulose – Sigma Chemicals Medium Fibrous
- Hydroxy Butyl Methyl Cellulose – Sigma Chemicals
- Flour – rice, maize, wheat, soy, rye and corn flour commercially available grocery lines

All of the above matrices can be used for lubricants where the levels of metals are much higher. However, the following are particularly useful choices of matrices for blood and other body fluid analysis, which can also be used for analysis of lubricants or water samples.

Aluminium hydroxide $[Al(OH)_3]$: A very high quality aluminium hydroxide is produced in Western Australia. It is analytically relatively clean and cheap, and is being considered as a matrix.

Cellulose: Cellulose is an excellent theoretical matrix choice in that it is typically low in heavy metal concentration. A variety of ultra-pure cellulose was tested for compactability, wettability and metal content. The physical characteristics of cellulose as such (it was the original matrix) make it important material as a potential matrix. Particularly useful is fibrous cellulose in the form of cellulose filter papers (Whatman

540, but also 541, 542 and other cellulose filter papers, Whatman International Ltd, Maldstone, England).

Flour: Newly acquired rice flour has proved exceptionally robust under wetting and drying conditions and may also be advantageously used as a matrix.

5 In addition to simply using the matrix material as supplied, relevant matrices were leached and the leached residue tested to see if significant metals could be leached, thereby reducing the metal content of the matrix and possibly rendering it more useful by lowering the level of contaminant metals, or actually reducing the level of metals in the sample to a level where previously unsuitable material would now be suitable.

10 EXPERIMENTAL

(1) Chemical Characterisation

Solution ICP-MS: In order to assess the 'purity' of the respective potential matrices, appropriate sub-samples of water-soluble materials were dissolved in Milli-Q (mQ) water and made to volume. Water-insoluble samples, (primarily the inorganic
15 materials) were subjected to both cold and/or hot (or both) hydrochloric, nitric, aqua regia and nitric-hydrofluoric acid leaches. The leachates were recovered, made to volume, appropriately diluted and analysed by solution introduction ICP-MS. The leached residues were recovered and a selection of sub-samples subjected to total dissolution followed by solution ICP-MS analysis using a VG PlasmaQuad 3 ICP-MS
20 made by VG Elemental, Ion Path Road 3, Winsford, Cheshire CW7 3BX, United Kingdom. Further selected residue sub-samples, along with unleached equivalents, were subjected to total acid dissolution, made to volume, diluted and again analysed by solution introduction ICP-MS.

The solution experiments facilitated elimination of several of the potential matrix
25 candidates, having unacceptable concentrations of analytes of interest in the raw material and analytes little, or not adequately, reduced by acid leaching. The 'solution' assessment indicated that cellulose and aluminium hydroxide were the best candidates but that both of these may contain certain analytes of interest. Because of the need to dilute the solutions for ICP-MS analysis, very low apparent concentrations in solution
30 frequently translated to significant concentrations in the sample when corrected for mass and dilution; in many cases, these analytes may not be present or, if present, present at very much lower concentrations. To test this thesis, 'raw' sub-samples, and corresponding leached residues where applicable, were pressed into 'briquettes' (see below) and subjected to comparative qualitative UV LA-ICP-MS analysis.

35 **Laser Ablation ICP-MS:** It is not necessary that the sample matrix will contribute an equivalent amount of material to the analytical sample as the blood or other fluid.

The incorporation of the matrix and its ionisation will not be equal to that for the blood contained in it. Because of this, the contribution of matrix to the analytical signal will not necessarily be in proportion to its relative matrix/blood ratio. Hence, it was necessary to determine what relevant contribution the matrix has to the analytical signal during a real analysis. Laser ablation analysis of the matrix was therefore also undertaken. Because the use of argon as a carrier gas is the traditional method of transport of ablation debris to the plasma this was the initial gas used for all experimental purposes. However, helium is finding an increased following in the scientific community as a transport gas as it often gives improved sensitivity and reduced isobaric interferences. Consequently this gas was also investigated.

(II) Physical Characterisation

Physical characterisation of potential matrix materials included assessment of compaction integrity, both at 500 and 1000 kg/sq in, wettability to blood and aqueous solutions, integrity after sample addition, contrasting behaviour of single and multi-component matrices, and internal standard introduction. Results from some of these investigations are detailed below.

The use of an internal standard is necessitated because of the variability in ablation efficiency between samples. There is no way of controlling the "fluence" variation (variation in the efficiency of coupling and hence power transfer of the laser energy to the sample) from sample to sample. Because of this, varying amounts of analyte will reach the plasma depending on the relative fluence between samples. Consequently, it is necessary to ensure that there is a mechanism for estimating the amount of material being transported to the plasma for each sample. The method used for an infrared laser was to measure the scattering of light by the transported particles. However, this mechanism is not possible when a UV laser is used (the laser used for these experiments was a frequency quadrupled Nd-YAG UV Microprobe Laser System operating at 266nm in pulsed Q-switched mode. The Laser System was manufactured by VG Elemental, Cheshire, United Kingdom.

However, spiking a simple element cocktail into the matrix, either prior to, or concurrent with, sampling provides a useful and inexpensive internal standard for quantification experiments.

RESULTS AND DISCUSSIONS

Details of eighteen experiments completed during the period October-December 2002 are set out below. Sixteen of the experiments relate specifically to physical and chemical characteristics of the matrix, and analysis of absorbed aqueous standard, mineral CRM and blood samples. The remaining two experiments, Experiments 13 and

15, deal with the analysis of oil samples – these are reported together at the end of this section.

The resulting analytical data is presented in a series of Appendices identified by experiment number, for example, 'Appendix Experiment 12'. These appendices should
5 be viewed in conjunction with the relevant commentary on the individual experiments as contained herein. Frequently, averages of data and % standard deviations (coefficient of variations) have been computed.

In most appendices, isotopic data has been computed to 100 per cent elemental concentration using natural isotopic abundance relations. In a small number of cases,
10 data is presented solely as isotopic concentrations at the measured isotopic mass. This is clearly indicated in the respective appendices.

In an attempt to optimise signal response, peak hopping instead of normal scanning acquisition was employed. Under this analytical regime, data acquisition at each isotopic mass occurred on three channels only. Not uncommonly, transient
15 electronic spikes may be recorded on one of the three channels. The on-board computer processes the data from all three channels and reports the results as raw count 'concentrations'. Where a measurement includes a transient spike, the resulting raw counts for that analyte may be considerably elevated relative to duplicate or replicate analyses of the equivalent analyte in the same sample. This leads to often-
20 marked concentration contrasts for specific analytes in these samples. The problem may be overcome by increasing, to say seven, the number of channels over which individual isotopic mass data is collected. Under these circumstances, a normal 'smoothing' algorithm may be automatically applied across the seven channels to produce precision results for duplicate or replicate analyses. Having established this as
25 being a major cause of analyte variability, analytical protocols have been appropriately modified to allow data collection over the increased number of channels.

Another cause of analyte variability may be due to possible surface 'contamination' of the collection matrices. To minimise contamination, the top pad of a matrix wad has been removed so that there is no airborne contamination on the surface
30 to be analysed. In an embodiment of this process, the matrix pads are prepared in a sterile, dust-free clean room, enclosed in a container which may only be breached immediately prior to sample collection. Improved analytical precisions, following implementation of this protocol, are attributed to the sample preparation

Correction of data for identified transient spikes had led to a marked
35 improvement in analyte reproducibility and, hence, 'precision' data.

Example 5: Matrix And Blood-Related Experiments**Experiment 1**

The aim of this experiment was to develop and test procedures to produce 3 mm diameter test tablets as a prelude to physical characterisation of sample matrices. For this purpose, an XRF pressed powder vacuum press was modified, and new dies manufactured, to facilitate pellet production. Matrix materials chosen for the inaugural production tests were glucose, cellulose and a 1:1 mixture of the two; initial compaction pressure was 500kg/sq in. Initial physical and chemical investigations were undertaken concurrently until preferred matrices were identified.

Pelletising of glucose required the use of weighing paper between sample and metal on the press die. Absorption of liquid appears good.

Cellulose pelletised quite well, with very good strength. However, fluid absorption was slow. A 1:1 mixture of glucose and cellulose powder pelletised well without the need for weighing paper between pellet and die. Pellet strength was improved over glucose alone and fluid absorption was intermediate between rates for glucose and cellulose powder pellets compacted at equivalent pressure.

Experiment 2

The principal objective in this experiment was to assess the chemical purity of a range of potential matrix materials. Sample preparation for analysis was undertaken concurrently with pelletising press modifications. Various matrices, including pig-toe mussel shell, glucodin, glucose, cellulose, hydroxy butyl methyl cellulose (HBM cellulose), TiO_2 and $\text{Al}(\text{OH})_3$ were leached, dissolved or digested in preparation for solution ICP-MS purity assessment.

Method

Pig toe mussel (Sample A, B, C and D) - ~1.5g pearl seed taken, dissolved in 20mL 1:1 HCl:mQ water, then taken to dryness. 4mL of HNO_3 :mQ 1:1 added, heated and made up to 100mL with mQ water. Diluted x20 with mQ (2ppb Ir, Rh) water for ICP-MS.

Glucodin (Sample E and F) + Glucose (Sample G) - ~1.5g Dissolved in 100mL of mQ water. Diluted x5 for ICP-MS.

Cellulose (Sample H) + HBM Cellulose (Sample I) - ~0.5g digested in 20mL CHNO_3 for 36 hours, reduced to 10mL and made up to 100mL with mQ water. Diluted x5 for ICP-MS.

TiO_2 (Sample 001) + $\text{Al}(\text{OH})_3$ (Sample 003) - Leached with 1:1 HCl:mQ water for 36 hours, decanted and washed 3 times with mQ water (~20mL). Decanted solution (leachate) made up to 100mL with mQ water. Diluted x10 for ICP-MS.

TiO₂ (Sample 002) + Al(OH)₃ (Sample 004) - Leached with 1:1 HNO₃:mQ water for 36 hours, decanted and washed 3 times with mQ water (~20mL). Decanted solution (leachate) made up to 100mL with mQ water. Diluted x10 for ICP-MS.

Residues were dried and saved for LA-ICP-MS.

5

This experiment was concerned with the determination of the trace element concentrations in prospective matrices for blood (and other fluid) collection, together with looking at some of the results of leachates of titanium dioxide and aluminium hydroxide.

10 The results for the leachates are detailed (Appendix Experiment 2). It may be possible to indicate that aluminium is obviously leached from the aluminium hydroxide matrix, but also from the titanium dioxide matrix, and conversely titanium is leached from the titanium dioxide matrix and there is also some indication of leaching of titanium from the aluminium hydroxide matrix. In the case of titanium dioxide, HCl appears to be
15 more aggressive than HNO₃, whereas the reverse is the case for the aluminium hydroxide. Concentrations of manganese, copper, strontium, zirconium are found from the leachates of both matrices while zinc, rubidium, barium and lead appear to be quite concentrated in leachates from the titanium dioxide matrix. In the aluminium hydroxide matrix tin, gallium, zirconium, hafnium and uranium appear to be present in leachates
20 from this matrix.

Total digest and/or solubilization data of pig-toe mussel, glucodin, glucose, cellulose and HBM cellulose are also presented in Appendix Experiment 2. The pig-toe mussel contains significant concentrations of lithium, aluminium, titanium, manganese, copper, zinc, rubidium, strontium and barium. While this would imply that the matrix is
25 not suitable as a blood collection matrix, because of the concentration of these elements, it is also necessary to analyse the pig-toe mussel material with sample attached under laser ablation conditions rather than solution conditions to make sure that these elements are also carried over by laser ablation and not just present in total digests. In the case of glucodin, glucose, cellulose and HBM cellulose all contain
30 significant amounts of aluminium, titanium, chromium, manganese, nickel, copper, zinc, rubidium, strontium and barium while cellulose matrix alone, in addition to containing these elements, also contains significant concentrations of lead and bismuth; both cellulose and HBM cellulose also contain concentrations of zirconium, tin, thallium and thorium not found in the glucodin and glucose.

35 Although these matrices all contain significant amounts of trace elements in the ppb range, this does not necessarily preclude them from use as a sample collection

matrix as conventional blank correction can be used to overcome problems associated with blank content. This can be further emphasised by the fact that Inter-element ratios could be used to determine, and to augment, blank corrections by looking at relationships between metals and tracing these through to the final analytical protocols

5 Experiment 3

The purpose of this experiment was to further test, the pelletising and adsorption characteristics of cellulose powder, glucose, and starch, and mixtures thereof, and to check the dissolution/absorption characteristics of the pellets by SY-2 (mineral CRM, , Canadian Certified Reference Material Project (CCRMP), Table 1 solution. The results of Experiment 3 are set out in Appendix Experiment 3

Cellulose powder alone works well. The glucose undergoes surface dissolution leaving holes on the surface. The starch absorbed water and expanded, causing the surface to bulge. Under the pelletising pressure of 500 kg/sq in, the cellulose powder is tightly compressed and it takes some 10 to 15 seconds for fluid absorption. This suggests that a more fibrous cellulose with an 'open' structure may be preferable. To this end, further experimentation with fibrous cellulose is indicated. In addition, further experimentation with powdered cellulose at differing packing pressures is warranted.

Experiment 4

The aim of this experiment was to assess the absorptivity and mechanical stability of cellulose powder pellets compacted under differing pressures. In the first instance, powdered cellulose was suspended in mQ water and vacuum filtered. The collected filter cake was mechanically incoherent. This caused it to flake and fall apart. However the adsorption of solution was rapid.

Cellulose powder compacted under a pressure of 100kg/sq in, while mechanically robust, still absorbed slowly. At low compaction pressure, estimated to be about 50kg/sq in and achieved by turning the tightening screw on the press just until there was resistance, the resulting pellets illustrated rapid absorption. Furthermore, the pellet holds together well. The experiment appears to confirm that compaction destruction of porosity rises with increasing pressure thereby rendering the matrix progressively less absorptive.

Experiment 5

The aim of this experiment was to quantitate trace elements in a blood sample using internal standards. The experiment also tested the absorption of SY-2 (mineral CRM) and blood onto cellulose pellets, robustness of the doped pellets when subjected to LA-ICP-MS analysis, assess levels of possible contaminants, evaluate results arising from the doped matrices and assess the comparability between 'wet' and 'dry' matrices.

The following instrument settings were used: Lens voltages – Lens 1, 2, 3, and 4 respectively –10.8, -22.6, 0.7 and –13.3 Volts, Collector – 4.6 Volts and Extraction, -332 Volts; Gas Flows – Cool gas 13.6 L/min, Aux gas 0.81 L/min Neb gas 0.74 L/min and Oxygen gas 0.00 L/min; Torch box positions – X, Y and Z axes respectively 932, 165 and 250 steps; Multiplier voltages – H.T. pulse count –2634 Volts and H.T. analogue) 5 Volts; Miscellaneous settings – Pole bias –2.2 Volts, R.F. power 1500 Watts, Perl speed 0%; PlasmaScreen is OUT, S-Option pump is OFF.

10 Samples of blood were obtained from a subject with the aid of a SoftTouch lancet device (used for home blood glucose testing and manufactured by Boehringer Mannheim, Germany) applied to a pre-cleaned (absolute ethanol wiped) area of a fingertip. Successive drops of blood were encouraged to form through application of pressure. The drops were directly 'touch' applied to 3mm diameter by 2mm deep sample collection matrix tablets formed by pressing granular cellulose (Sigma 15 Chemicals Microgranular powder) under a load of 500 kg/sq. in. The matrix tablets were affixed to a Perspex disc, 37.5 mm in diameter and 6mm deep, fabricated from Perspex rod, using 3M Scotch Permanent Double Stick Tape. The volume of the drops was estimated to range between 30 and 70 microlitres. No preservatives or anticoagulants were used and there was no requirement to store the blood prior to 20 application to the collection matrix, or subsequent analysis. However, there is provision for loaded sample collection matrix tablets to be refrigerated and stored following oven drying at 60°C for one hour.

Four blood samples were prepared; two were oven dried and two were maintained "damp". Duplicate sets of equivalent SY-2 CRM-doped (Syenite, Canadian 25 Certified Reference Material Project) matrix pellets were prepared by pipetting 50 µL of the standard solution onto the respective matrix tablets and drying thereby generating matrix matched standards. The SY-2 CRM contains calcium, iron, magnesium, potassium and so forth and this provides a high ion flux that is possibly equivalent to the ion flux expected of blood. Hence, any ion effects that were taking place would be 30 comparable in the blood and SY-2, as compared with a straight aqueous standard solution.

The sample holder, with affixed blood- and CRM- doped matrices was placed into the laser ablation cell of the UV Microprobe Laser System attached to a VG PlasmaQuad 3 ICP-MS both manufactured by VG Elemental, United Kingdom. The 35 laser is a frequency quadrupled Nd-YAG operating at 266 nm; 10x10 matrix raster

ablation of the samples was undertaken in pulsed Q-switched mode at a fluence of 6.2 millijoule for 60 seconds.

The output data was acquired as raw counts from on-board software and exported into Excel and manipulated. No algorithms were used for computations. The
5 raw count data for both blood and CRM samples were matrix blank corrected by subtracting the averaged matrix blank value from the individual blood and SY-2 values. From these corrected data % Standard Deviations were computed as a measure of precision. Finally, trace element compositions for the 11 analytes examined in the exemplary run were computed with reference to matrix matched SY-2 CRM values.

10 Data obtained is set out in Appendices Experiment 5A and 5B.

As indicated above, part of the experimental design was to determine whether it was necessary to fully 'dry' the sample prior to analysis. Collection of blood onto a matrix without the drying step as detailed above, may lead to a sample being slightly damp. Hence, it was necessary to determine whether variation in the moisture content
15 of the matrix would affect the readout of concentration of elements in the matrix.

Consequently two sets of samples of cellulose were set up and, in addition to 'wet' and 'dry' blood, SY-2 certified reference material doped samples were also prepared in an attempt to quantify the concentration of metals in the blood. Blood samples and SY-2 were spiked onto cellulose in duplicate and one set of blood samples was analysed
20 'wet'. A second subset was taken and dried (as above) and the samples were analysed dry. Data from these experiments is also presented in Appendix Experiment 5A

Following analysis, results for the wet samples were blank corrected and data produced. Simple inspection of the data for the 'wet' blood samples indicates relatively high variability in analyte concentrations particularly in the case of lead and zinc where a
25 variation of $\pm 100\%$ is recorded. The analysis of SY-2 certified reference material is far more uniform.

For the dry sample, the results are better. Reproducibility is improved and results are more uniform. From the blank corrected values for the dried blood sample it can be seen that, with the exception of barium, the results are meaningful. Barium
30 results go negative and this is probably due to the fact that the barium signal is small relative to the blank – the blank is quite high. However, both lead and zinc are much improved and, if these are used to calculate concentrations of these elements in the blood, based on SY-2 concentrations (calculated in Appendix Experiment 5B) the blood values and expected blood values from the literature are quite close for the analytes
35 under consideration. SY-2, a certified reference material, has been used for a number of reasons. First, use of simple aqueous solution on the collection matrix would not, on

ablation, have provided a significant ion flux. The SY-2 contains calcium, iron, magnesium, potassium etc (see Table 1) and this provides a high ion flux that is possibly equivalent to the ion flux of the blood. Hence, any ion effects that were taking place would be comparable in the blood and SY-2, as compared with a straight aqueous solution. Thus a normal CRM, that has a relatively high matrix concentration will suffice.

The above experiment, including instrument settings and internal standardisation as described, is equally applicable to simpler biological fluid samples such as components of whole blood (eg. serum or plasma), urine, sweat, tears, cerebrospinal fluid and the like. The sample collection, handling and analysis of such fluids is simpler and thus greater accuracy can be achieved.

Experiment 6

This experiment was conducted to analyse the titanium dioxide and aluminium hydroxide matrices, both before and after leaching (leached residues from Experiment 2). The data produced in this experiment ties in with the leachate data from Experiment 2. Upon total dissolution, solutions derived from titanium dioxide have very high concentrations of titanium, while those derived from digestion of aluminium hydroxide are similarly rich in aluminium. Accordingly, these two elements have not been measured.

The purpose of the experiment was to evaluate the efficacy of acid cleaning of the white oxide matrices. Hence, appropriate sub-samples of 'raw' titanium dioxide and aluminium hydroxide, together with their hydrochloric- and nitric acid-leached equivalents, were digested in a sulphuric/hydrofluoric acid, made up to volume, diluted and analysed by solution introduction ICP-MS. The leachates derived from HCl- and HNO₃-leaching of bulk titanium dioxide and aluminium hydroxide were analysed in Experiment 2 and the results reported in Appendix Experiment 2.

The comparison of the "raw" original material and the HCl- and HNO₃-leached residues show that, for titanium dioxide, its HCl-leached residue and associated leachate, weak to strong leaching of lithium, manganese, copper, zinc, gallium, rubidium, strontium, (zirconium), barium, lead, (thorium) and uranium has been achieved. Here, there is generally a good mass balance between concentration in the original versus the sum of concentrations in the leachate and leached residue. In contrast, concentrations of vanadium, chromium, nickel, germanium, yttrium, zirconium, niobium, tin, antimony, hafnium, tantalum and tungsten in the raw material are unaffected by HCl-leaching.

For titanium dioxide, its HNO₃-leached residue and associated leachate, weak to strong leaching of lithium, (chromium), manganese, copper, zinc, gallium, rubidium, strontium, (zirconium), barium, lead and (thorium) is evident. In contrast,

concentrations of vanadium, (chromium), nickel, germanium, yttrium, niobium, tin, antimony, hafnium, tantalum, tungsten, (thorium) and uranium are little or unaffected by HNO_3 -leaching.

Turning to the aluminium hydroxide matrix, HCl and HNO_3 both have a similar
5 leaching response with both acids weakly to strongly leaching all elements occurring in significant concentrations in the aluminium hydroxide matrix. The elements involved are lithium, beryllium, chromium, manganese, copper, gallium, strontium, zirconium, tin, hafnium, thorium and uranium. Hence, use of these acids to pre-clean the matrices is recommended. Both can be leached quite easily in both HCl and HNO_3 .

10 Of particular importance is the presence of gallium in the aluminium hydroxide matrix. A small amount is acid-leached but this does not impact its potential of being used as an internal standard; the same holds true for zirconium. Although not as high as zirconium in the titanium dioxide matrix, zirconium in aluminium hydroxide could still be used for a double internal standard based on gallium and zirconium. There is a
15 possible problem with the aluminium hydroxide matrix in that there is copper in it but the copper tends to be relatively uniform and if copper results in previous analyses are considered, reasonable results for copper are obtained by doing blank corrections. It should be remembered all the time that although these metals are present in the matrix, they may not contribute an equivalent amount to the determination of metals in blood
20 because they are not transported as much as the blood to the plasma. The blood tends to fill interstices and sit on top of the matrix; hence, these elements may not contribute a significant amount to the concentrations that are present in analysed, so-called blood.

This experiment demonstrates that it is possible to variably reduce and/or eliminate a range of trace elements from titanium dioxide and aluminium hydroxide
25 matrices. When combined with previous experiments, it would appear that possibly two matrices, aluminium hydroxide and cellulose, may constitute particularly suitable matrix materials.

Experiment 12

The purpose of this experiment was to examine the efficacy of a fibrous
30 cellulose mat (Whatman 540 filter paper, Whatman International Ltd) as a sample collection matrix. This material is an efficient absorber of fluids, but its 'coarse' fibrous texture may result in variable ablation characteristics. Six duplicate sub-samples of the cellulose mat were taken and pre-prepared as follows: Two duplicate sets were rinsed for 10 minutes with 50% aqua regia and dried; a further two duplicate sets were washed
35 overnight in aqua regia and dried while the remaining duplicate sets were left unwashed. One set each was doped with 2ppm multi-element standard and dried whilst

the second set of each was retained as blanks. It was observed that the fibrous cellulose mat, rinsed for 10 minutes with aqua regia, upon drying was rendered 'harder' than the other two (unwashed and overnight washed) mats.

The blanks and doped equivalents were analysed by LA-ICP-MS and the results of analysis are recorded in Appendix Experiment 12. Upon ablation, it was observed that for the 'hardened' rinsed matrix, the laser penetrated through the whole mat, whereas for the other two, the laser did not penetrate all the way through. This observation clearly implies that the contrasting physical characteristic of the fibrous cellulose mat impact upon laser penetration and, hence, lasing characteristics. With reference to the relevant Appendix, pages Experiment 12/3 and 12/4, it is clear that, for cerium-normalised data, data for the 'hardened' rinsed fibrous cellulose mat, which exhibited complete laser penetration, gives rise to the best overall precision data. Indeed, most analytes have precisions of less than 10% and frequently less than 5%. This outcome further emphasises the potential value of fibrous cellulose as a matrix material.

Experiment 16

The objective of this experiment was to evaluate potential sensitivity improvements for aqua regia and ammonium fluoride (NH_4F) doped 3:1 $\text{Al}(\text{OH})_3$:cellulose matrices.

From a 3:1 $\text{Al}(\text{OH})_3$:cellulose mixture, six triplicate sets of pressed pellets were prepared. These unwashed triplicate pellet sets were affixed to a Perspex disc. One set was left 'blank' and a further set was doped with 1ppm multi-element standard; both were oven baked. Two of the remaining four triplicate sets were doped with 5 μL of 50% aqua regia and oven at 105°C for 2 hours; the remaining two triplicate sets were doped with 5 μL of 1M ammonium fluoride (NH_4F) and oven baked. One set each of the aqua regia and ammonium fluoride treated pellets were further doped with 1ppm multi-element standard and dried.

A further sample of the 3:1 $\text{Al}(\text{OH})_3$:cellulose mixture was washed with aqua regia, rinsed and dried. This material is referred to as the washed matrix. From this washed matrix, equivalent triplicate sets of pellets were prepared as for the unwashed matrix described above. It was observed that the 50% aqua regia doped matrices were not as mechanically robust as other matrices prepared in this experiment. All triplicate sets were analysed by LA-ICP-MS. The results for the unwashed matrices are presented in Appendix Experiment 16A while those for the washed matrices comprise Appendix Experiment 16B.

When results for unwashed material, that is, no aqua regia wash, are considered, it is apparent that the results are significantly better for unwashed, than for the washed, material. For blank corrected matrices, normalised to cerium, precisions for the unwashed material are better than those of the washed matrix. This outcome suggests that there is no fundamental need to wash 3:1 $\text{Al}(\text{OH})_3$:cellulose matrix.

Disregarding, the blank corrected, cerium normalised data for the present, and considering only the 'raw' 1ppm doped matrix data, the recorded precision measurements for both unwashed and washed matrices show a general improvement in the NH_4F doped matrices. This apparent improvement in sensitivity may result from improved ablation of the matrix possibly through production of a more volatile atmosphere in the presence of NH_4F .

Experiment 18

The several previous experiments have sought to identify appropriate clean matrix materials together with preferred compaction, absorption, ablation and pre-treatment characteristics. Particularly preferred matrix and analytical conditions for most test samples, and particularly useful for blood and other body fluid samples, were identified as Whatman 540 filter paper, ablated at 10Hz at a fluence of between 4 and 9 Millijoule with a flow of argon between 900 and 1000mL per minute.

In the course of this work, consideration was given to the question as to whether it may be possible to prepare a blood sample in such a way that it was matrix supported, rather than matrix absorbed. If this could be achieved, then it may be possible to ablate blood samples free of matrix. In this way, analytes present in the analysis would be derived from the blood alone. Consideration of direct analysis of supported, rather than matrix-absorbed blood, arose from the observation that, during the experimental procedures segregation of blood serum and plasma appeared to occur. The observed probable segregation was not considered to be a significant problem; the laser ablation protocol was designed in such a way that the laser would penetrate through any dispersion front in the matrix, thereby sampling any segregated blood and consequently 're-assembling' or re-combining the analyte cocktail. Nonetheless this observation suggested that it might be possible to overcome any potential matrix interference by ablating only dried blood.

It was reasoned that if a shallow, 3mm diameter, 125 micron deep, depression was cast into the surface of the matrix pellet, then a drop of blood delivered to the depression would flow to fill the depression and present a flat surface away from the depression lip (meniscus) for subsequent lasing. A requirement would be that no chromatographic segregation of serum and plasma occurred. To this end, it was further

reasoned that if the 3:1 $\text{Al}(\text{OH})_3$:cellulose powder was compacted under high pressure (at least 1 tonne/sq in), then the matrix may be rendered effectively impervious and simply support blood as it coagulated and dried.

Consequently, a new die for the vacuum press was fabricated to produce a 6mm diameter pellet into which was impressed a 3mm diameter by 125 micron deep, flat bottomed circular depression. An appropriate number of new pellets were pressed at 1 tonne/sq in pressure.

Micro-lltre samples of blood were delivered to, and contained within, the surface depressions on the surfaces of ten matrix pellets; five of these pellets were air dried at ambient temperature and the remaining five oven dried at 60°C. A further two blood drops were applied to the Perspex mounting disc and dried. Here, the surface of the dried blood drops was not flat, but rather, strongly undulating.

On application, it was clear that some plasma segregation and absorption occurred, causing a volume increase and expansion in the tightly compressed cellulose powder. However, the pellets retained sufficient mechanical integrity to allow LA-ICP-MS analysis. When ablated, the 'serum' tended to fragment in 'chunks' giving rise to somewhat variable results. Notwithstanding, the counts obtained were reasonable for most elements.

For the matrix free blood drops, dried onto the Perspex support, the ablated blood was far more coherent, with nice ablation. However, as noted above, the surface was strongly undulating leading to changed laser focal conditions and, hence, non-optimal results.

Given that the aluminium hydroxide:cellulose matrix was not impervious, the matrix free approach described above can be adopted, i.e. use impervious substrate, such as Perspex, into which 3mm diameter by 125 micron deep circular impressions have been pressed, moulded or machined. Each sample collection device can contain two such depressions, one for a matrix-matched, trace metal-doped standard reference blood, and the second to contain and confine the unknown blood sample. Alternatively, a matrix-matched, trace metal-doped reference blood could be inserted into the analytical run such that each unknown had a standard immediately adjacent to it. This would lead to 33% reference samples in the analytical run as opposed to 50% if standard and unknown were applied to the same collection device.

The results from this Experiment are presented in Appendix Experiment 18. This experiment examined heat and air-dried blood partially absorbed into an aluminium hydroxide:cellulose powder matrix, and matrix-free blood dried onto an impervious Perspex substrate.

If the corrected and normalised "no-matrix" blood is examined, the numbers are reproducible. Indeed, values are commonly comparable to the dried material. In the 'no matrix' blood, both mercury and lead are recorded and the reproducibility of lead is with a precision of 14%. Good numbers are also recorded for uranium on the dried material, but in the blood matrix alone, the numbers are considered to be 'below detection limit', consistent with a matrix uranium background and anticipated absence in the blood.

Example 6: Wear Metal Analysis In Oils

Experiment 13

The objective of this experiment was to carry out pilot analysis of wear metals in engine oil. It is held that the technology being investigated is equally applicable to the analysis of wear metals in oils, and that wear metals analysis is a major global industry aimed at early detection and prevention of catastrophic plant failure. Such early detection is of particular importance to the military, airline, shipping and mining industries where component failure (automotive, heavy machinery, weaponry and the like) may lead to tragic loss of life and destruction of expensive plant.

Oil from the engine of a 'new' Ford Fairlane was sampled hot, with the engine still running, via the dip-stick. Oil from a single dip of the dip-stick was transferred to both an unwashed and washed 3:1 Al(OH)₃:cellulose powder matrix pellet pressed at 500kg/sq in. Duplicate pellets (without oil) were prepared as blanks and all four pellets analysed by UV LA-ICP-MS. Instrument settings as for Experiment 5 were used, with minor adjustments for day-to-day variations. The results of analysis are presented in Appendix Experiment 13.

When blank corrected, there is very little difference between results obtained on the unwashed and washed matrices. If the two matrices are treated as a single matrix, then precisions, with the exception of Iron, are excellent, commonly being <1 for the restricted range of analytes expected in oil. Reproducibility of the data, are thus excellent and this is graphically illustrated in the X-Y log plot of 'concentration' versus elements comprising Chart Experiment 13/1. Here, consistent with the precision/reproducibility data, Iron excepted, the two profiles are effectively superimposed upon each other.

The experiment clearly indicates the general reproducibility of the analysis and indicates considerable promise for the technique.

Experiment 15

This experiment had as its main objective, the analysis of oil from the engines of five different cars, collected under the same conditions as described above, that is hot

with the engines running, on three consecutive days, to assess whether contrasts in wear metal content in oil from cars of contrasting age, engine capacity and, presumably oil used, could be established. For one 'old' car, which required frequent oil top-ups between services, a sample of the new top-up oil was available for comparison. The oil
5 was collected as for Experiment 13, but in duplicate on unwashed 3:1 $\text{Al}(\text{OH})_3$:cellulose powder pellets pressed at 100kg/sq in pressure; new reference oil was dipped with a glass rod and applied, in duplicate, to equivalent pellets. All samples were analysed by UV LA-ICP-MS; the results of the expanded range of analytes are presented as Appendix Experiment 15.

10 During the course of the analysis, eleven glass standard measurements were made. The precisions on the raw glass data are generally in the range 10 to 20%. However, when the raw data are normalised to average cerium, precisions are generally excellent and, with the exception of selenium, cadmium and mercury, are <10 ; selenium and cadmium are just marginally higher and mercury sits at 24%. The cerium
15 normalised glass standard data have been plotted in a log X-Y line chart plot which comprises Chart Experiment 15/1. Here, it is clear that the several profiles essentially superimpose, consistent with the very good precisions and reproducibility. In addition to the glass standard, 10 air blank measurements were made throughout the analytical run. These have been drift corrected and the average drift corrected air blank has been
20 used to correct the reported data.

Assessment of the data clearly demonstrates significant, and often marked differences, in specific analytes between the engine oils from the different vehicles. Oil from two cars, 'John' and 'Scott', were selected to demonstrate these contrasts. 'John' engine oil is plotted as a log X-Y line chart in Chart Experiment 15/2 while 'Scott' oil
25 comprises Chart Experiment 15/3. Examination of the respective Charts illustrates that while, there is general profile superimposition for the respective replicate oil analyses, there are some clear difference in the shapes of the respective profiles as well as peak height contrasts between equivalent analytes. Chart Experiment 15/4 graphs the averaged composition of 'John' and 'Scott' oil ($n=6$). This latter Chart clearly
30 emphasises the marked compositional contrast between the two oils. Hence, from this experiment, it may reasonably be concluded that the technique can readily identify and measure analyte contrasts in the examined engine oils. It is clear from the pilot experiments that wear metal analysis of oils of plant in service by LA-ICP-MS techniques is feasible and useful. The experimentation into the analysis of wear metals
35 in oils indicates considerable potential economic benefits of being able to, for example, regularly monitor potential component wear, through 'dip-stick' sampling, in plant in

service, that is without the need to plant take off-line, are large. In this way plant down-time can be carefully scheduled with minimal impact upon operations.

The use of a defocused laser to ablate sample matrices is a variation of the protocols described, which can be used to improve laser coupling to the sample. If a laser is focused on the surface of a sample, the first crater it produces is a response to the laser focal point being on the surface of the sample. As soon as the surface material has been ablated and removed, the next ablation event (laser shot) is into the crater area from the first shot where there is no focus and, therefore, the laser coupling is diminished. If, however, the laser is focused below the surface, that is, it is defocused at the surface, potentially it is now possible to generate a more active ablation because a large amount of material can be ejected from the middle of the sample because the focussing is below the surface. Hence, it might be expected that at least the first and second shots will produce a lot of ablation debris and therefore this may increase the sensitivity because, at this stage the ablation ejecta is a powder/aerosol and this may be more efficiently transported to the plasma torch. For the existing equipment, laser defocusing can be fairly readily achieved manually. Modern lasers have automatic defocus capabilities where the depth for defocusing can be simply programmed.

As a further modification of the present protocols, triple shot ablation, as compared with double shot, at each point in a 10 point by 10 point raster grid, may be used.

Example 7: Quantitation using solution doped matrices (further experiments)

In this example three fibrous cellulose matrices, being Whatman 541, high purity Whatman 541 and old Whatman 540 filter papers (Whatman International Ltd, Maidstone, England), were prepared as blank material by affixing to a support substrate using a backing tape; a sample of the backing tape (3M Scotch Permanent Double Stick Tape) was also analysed. The raw count data was analysed firstly as isotopic concentrations for the designated elements and secondly as elemental abundance concentrations derived from the isotopic data using natural abundance relations. All elemental data has been air blank corrected. Air blank correction has produced negative values for isolated analytes implying that the analyte concentrations in the average air blank are significantly higher than in the matrices for those analytes. Examination of the data illustrates generally high analyte air blank values.

All elements have been spike corrected (ie. normalised to an average value for the spike) and 'old' refers to fibrous cellulose substrates that have previously been opened and exposed to the laboratory environment through 'open' long-term storage. 'New' refers to sealed fibrous cellulose substrates opened for this experiment. With

respect to the single versus multiple layer substrate data, it appears probable that analysis of single layer substrates may have involved laser penetration into the backing tape. Hence, data for single layer substrates may reflect composite data whereas for the multiple layers, where the top layer was peeled off immediately prior to analysis, the data reflect only the cellulose matrix substrate.

The data illustrated lower concentrations for a significant number of analytes in multiple, relative to single, layer matrices; other analytes are essentially equivalent while some are higher. For many analytes, for example Cu, Zn, Sn, concentrations in the backing tape is very much greater than in the both the single and multi layer matrices but, here, the single layer matrices are much higher in these elements than the equivalent multi layer material. This strongly suggests that laser penetration to the backing tape has occurred and that much of the difference between single and multi layers has little to do with handling contamination.

Furthermore, the corresponding data for 'new' versus 'old' clearly demonstrates significantly lower overall concentrations in the new matrices, both single and multiple. This latter observation strongly suggests that long-term exposure of matrices to the laboratory environment has led to variable, but significant ambient laboratory contamination of exposed matrices.

Further experiments examined white and black Whatman 540 filter paper cellulose matrices (Whatman International Ltd, Maidstone, England) doped with 1ppm multi-element standard (details are provided in the table) and with blood.

The data have been matrix blank corrected. For many of the analytes the air blank is high and similar to the concentrations measured in the white and black cellulose blanks (matrices without samples applied).

The isotopic data, as obtained, was converted to elemental concentrations and the multi element standard and blood doped samples have effectively been doubly corrected. The respective white and black cellulose matrix blanks have first been air blank corrected using the average of two air blanks. Following this, the averaged data, for multi standard and blood doped white and black cellulose, have been corrected using the respective corrected air blank corrected white and black cellulose matrix blanks. There is good correlation between the averaged corrected values for white and black multi element standard doped matrix samples and white and black blood doped samples. Little difference exists between the multi element standard and the blood on white and black matrices. The data obtained in this experiment also illustrates excellent reproducibility for the vast majority of analyt across the mass spectrum in both multi element and blood doped matrices.

Comparison of the computed concentrations in the blood may now be compared with anticipated concentration ranges from the literature. Data for Fe, Cu, Zn, Sn, Ba and Pb show very good agreement.

Hardware optimisation

5 This experiment was to evaluate hardware optimisation at low, medium and high mass, using respectively manganese, lanthanum and lead. The isotopic data (isotopic concentrations), as obtained, has been rearranged and treated in a manner analogous to that in Example 7. For the current data, air blank, 540 matrix blank, 1ppm multi element standard and blood doped matrices were examined during optimisation at the
10 relevant masses. Again, the respective 540 matrix blanks have been air blank corrected by subtracting the averaged values from the averaged matrix blank values. Using the corrected matrix blanks, both the 540 multi element and blood doped matrices have been matrix corrected. Again using the corrected data, concentrations in ppb in blood have been computed.

15 The current data appear to indicate that low mass optimisation may be preferable. When doubly corrected, the indications are that, both for the multi element and blood doped matrices, optimisation at the lower mass, that is manganese, appears preferable to the mid mass and to the high mass. Once again, it is clear, with respect to quantification of trace element in the blood, matrix matched standards are of particular
20 value.

Detection limits and precision

The experiment was designed to establish detection limits, precision and quantitation for solution doped cellulose matrices. A series of standards were used for these experiments. In addition a reagent blank was also used.

25 Deionised water samples were doped, using a 'stock' multi-element standard solution, to produce a series of aqueous multi-element standard solutions with element concentrations of 100, 200; 500; 1000; 2000; 5000 and 10000 ppb. 100 μ L of each of these aqueous standard solutions was transferred to fibrous cellulose matrix pads, prepared from Whatman 540 filter paper (Whatman International Ltd, Maldstone,
30 England), using a pipette; the pads were affixed to Perspex supports using 3M Scotch Permanent Double Stick Tape. Deionised water matrix blanks were also prepared by pipetting 100 μ L of deionised water onto the matrix pads. In addition, solutions of three Certified Reference Materials, SARM's 1, 3 and 46 (South African Bureau of Standards) were diluted 250 times, and 100 μ L aliquots of each were doped onto Whatman 540
35 matrix pads. In all, 10 matrix pads of each aqueous standard concentration and CRM were prepared along with deionised water matrix blanks. A 2ppm samarium internal

standard solution spike was added to the respective matrix pads to facilitate internal normalisation; the spike was added using a pipette. All doped matrix pads were dried at 105°C for two hours prior to ablation.

Five of each set of ten prepared matrices were analysed on successive days.
5 The sample holders, with affixed matrix pads, were placed in the laser ablation cell of a UP 266 UV Laser System connected to an X Series ICP-MS with Xi Cone System (Thermo Optek (Australia) Pty Ltd, Rydalmere, Australia) and ablated on a 10x10 matrix raster using a UV laser operating at 266 nm, 10Hz at a fluence of 6 Millijoule and an argon flow between 900 and 1000 mL per minute for 60 seconds.

10 Samples were analysed manually and results have been corrected for air blanks, facilitating cross comparison between CRM and standard matrix matched samples. The output data was acquired as raw counts from on-board software and exported into Excel and manipulated. No algorithms were used for computations. From these corrected data, Standard Deviations and Coefficients of Variation have been
15 computed as measures of reproducibility and precision. Finally, quantitative trace element compositions for the 44 analytes examined in the exemplary run were computed for the CRM's; sub-20ppb detection limits for most analytes were achieved.

Data obtained data is set out in Appendix Experiment M1. It is also quite apparent that data for the standards, when plotted, indicate excellent calibration can be
20 achieved. Quantitation of data for the CRM's indicated extremely good agreement for elemental concentrations for all elements with values (for samples once diluted) in the optimum analytical range of the technique.

There are a number of points that this data demonstrates.

- 1) It is possible to achieve sub 5% precision for a wide range of elements using the
25 analytical protocols developed in conjunction with ICP-MS.
- 2) It is possible to achieve sub 20ppb detection limits for a wide range of elements simultaneously.
- 3) It is possible to achieve accurate quantitative data, using matrix matched certified reference materials, or other equivalent CRM's.

30 Examples of useful areas of application of the methods and devices of the present invention are:

- screening occupationally exposed workers for anomalous levels of a range of toxic metals;
- monitoring environmental exposure of the general population to toxic metals;
- 35 • screening populations for trace/ultra trace element deficiencies for preventative medicine

- screening trace/ultra trace element deficiencies, and toxic heavy metal excesses, in bloodstock, general livestock, zoo animals (including animals in endangered species breeding programs), and domestic pets for veterinary medicine; and monitoring heavy metal pollutants in slaughter animals for meat product quality control in the human food chain.
- Monitoring/detecting wear of mechanical components of plant, machinery and the like by analysing lubricating oils.

Although the invention has been described with reference to certain preferred embodiments, variations in keeping with the broad principles and the spirit of the invention are also contemplated as being within its scope.

APPENDIX EXPERIMENT 2

Element - ppb* In original	Li	Be	Al	Ti	V	Cr	Mn	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Rb
TiO ₂ /HCl -001 leachate	7	<1	8,340	174,555	<1	<1	435	<1	<1	457	364	8	<1	<1	<1	75
TiO ₂ /HNO ₃ -002 leachate	11	<1	13,780	76,451	<1	14	638	<1	<1	527	438	13	1	<1	<1	106
Al(OH) ₃ /HCl -003 leachate	37	4	41,530	180	<1	118	48	<1	<1	14	<1	2,357	<1	<1	<1	<1
Al(OH) ₃ /HNO ₃ -004 leachate	45	4	48,312	1,456	<1	17	33	<1	<1	50	<1	2,523	<1	<1	<1	5
Pig Toe A digest	63	<1	11,600	1,779	<1	<1	761,998	<1	<1	113	817	<1	<1	<1	<1	23
Pig Toe B digest	84	<1	9,956	2,086	<1	<1	475,395	<1	<1	138	890	<1	<1	<1	<1	43
Pig Toe C digest	109	<1	10,314	2,165	<1	<1	760,369	<1	<1	126	922	<1	<1	<1	<1	72
Pig Toe D digest	57	<1	9,424	1,922	<1	<1	966,818	<1	<1	170	421	<1	<1	<1	<1	59
Glucodin E solute	8	<1	2,378	91	<1	359	265	<1	107	18	149	<1	<1	<1	<1	20
Glucodin F solute	4	1	2,218	92	<1	327	208	<1	103	29	181	<1	<1	<1	<1	31
Glucose G solute	8	2	1,896	89	<1	345	96	<1	110	21	131	<1	<1	<1	<1	18
Cellulose H digest	9	7	22,353	1,391	50	798	296	<1	963	523	862	<1	<1	<1	<1	62
HBM Cellulose I digest	71	3	25,313	1,278	50	2,392	1,538	<1	1,282	1,671	1,413	<1	<1	<1	<1	78
* ppb in solution for leachates																

APPENDIX EXPERIMENT 2

Element - ppb* in original	Sr	Y	Zr	Hf	Mo	Ag	Cd	Sn	Sb	Te	Cs	Ba	La	Ce	Pr	Nd
TO2/HCl -001 leachate	134	<1	62	<1	69	<1	<1	<1	<1	<1	<1	2,808	6	8	<1	<1
TO2/HNO3 -002 leachate	195	1	180	<1	<1	<1	<1	<1	<1	<1	<1	3,250	8	11	<1	<1
Al(OH)3/HCl -003 leachate	170	<1	1,289	<1	<1	<1	<1	188	<1	<1	<1	<1	<1	2	<1	<1
Al(OH)3/HNO3 -004 leachate	189	<1	818	<1	<1	<1	<1	174	<1	<1	<1	<1	<1	3	<1	<1
Pig Toe A digest	237,704	<1	<1	<1	10	<1	<1	<1	<1	<1	<1	66,117	4	9	<1	<1
Pig Toe B digest	233,800	<1	1	<1	34	<1	<1	<1	<1	<1	<1	40,257	4	15	<1	<1
Pig Toe C digest	332,028	<1	<1	<1	41	<1	<1	<1	<1	<1	<1	85,251	8	16	<1	<1
Pig Toe D digest	303,598	<1	<1	<1	61	<1	<1	<1	<1	<1	<1	101,341	10	28	<1	<1
Glucodin E solute	188	<1	<1	7	63	<1	<1	<1	<1	<1	<1	72	1	2	<1	<1
Glucodin F solute	229	<1	<1	6	61	<1	<1	<1	<1	<1	1	43	<1	<1	<1	<1
Glucose G solute	22	<1	<1	<1	12	<1	<1	<1	<1	<1	<1	8	<1	<1	<1	<1
Cellulose H digest	357	<1	806	217	870	<1	<1	658	<1	<1	<1	168	6	12	<1	<1
HEM Cellulose I digest	13,800	<1	1,351	502	524	<1	<1	557	<1	<1	<1	480	6	11	<1	<1
* ppb in solution for leachates																

APPENDIX EXPERIMENT 2

Element - ppb* in original	Eu	Sm	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Hf	Ta	W	Hg	Tl	Pb	Bi	Th	U
TiO2/HCl -001 leachate	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	19,014	<1	4	10
TiO2/HNO3 -002 leachate	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	20,394	<1	3	<1
Al(OH)3/HCl -003 leachate	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	134	<1	<1	<1	1	<1	<1	3	135
Al(OH)3/HNO3 -004 leachate	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	131	<1	<1	<1	<1	<1	<1	2	152
Pig Toe A digest	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Pig Toe B digest	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Pig Toe C digest	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Pig Toe D digest	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Glucodin E solute	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	1	<1	<1	<1	<1	<1	5	1	<1
Glucodin F solute	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	5	<1	<1
Glucose G solute	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	41	<1	<1	<1
Cellulose H digest	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	24	186	137	55	<1
HBM Cellulose I digest	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	25	<1	<1	32	<1
* ppb in solution for leachates																			

APPENDIX EXPERIMENT 3

Sample	Sample No.	Pelletise	Absorption Rate of SY-2	Dissolution	Comments
Glucose	1	POOR	Fast	Yes	Pellet dissolved, absorbed quickly
Cellulose	2	OK	10-15 sec	No	Solution absorbed slowly
AR Starch	3	OK	Slow	Partial	Pellet swells
UR Starch	4	OK	Slow	No	Pellet swells
Glucose + Cellulose 1:1	5	OK	Slow	Partial	Absorption OK, partial dissolution, holes on surface
Glucose + Cellulose 3:1	6	OK	Slow	Partial	Dissolution of pellet
Cellulose + AR Starch 3:1	7	OK	V. Slow	Partial	Partial dissolution of pellet, holes left on surface
Glucose + AR Starch 1:1	8	OK	V. Slow	Partial	Dissolution and swelling
Glucose + UR Starch 1:1	9	OK	V. Slow	Partial	Dissolution and swelling
Cellulose + AR Starch 1:1	10	OK	Slow	No	Dissolution and swelling
AR Starch + Cellulose 3:1	11	OK	Slow	No	Dissolution and swelling
Cellulose + UR Starch 1:1	12	OK	Slow	No	Swelling of surface
Cellulose + AR Starch 3:1	13	OK	Slow	No	Swelling of surface
Cellulose + UR Starch 3:1	14	OK	Slow	No	Swelling of surface
UR Starch + Cellulose 3:1	15	OK	Slow	No	Swelling of surface
Glucose + Cellulose + AR Starch 1:1:1	16	OK	V. Slow	Partial	Dissolution and swelling
Glucose + Cellulose + UR Starch 1:1:1	17	OK	Slow	Partial	Dissolution and swelling

APPENDIX EXPERIMENT 5A

Isotope - Raw Counts	Mg 24	Ca 44	Mn 55	Fe 56	Cu 65	Zn 66	As 75	Se 77	Mo 98	Ba 138	Pb 208
WET											
"02/11/07 CELLULOSE AIRBL1"	38,010	14,080	2,719	25,180	2,686	377	660	432	138	111	
"02/11/07 CELLULOSE AIRBL2"	35,740	13,480	2,578	24,210	2,592	309	626	443	108	36	73
"02/11/07 CELLULOSE BLANK1"	60,150	24,560	7,263	888,700	15,140	8,261	671	328	1,542	5,132	8,886
"02/11/07 CELLULOSE BLANK2"	58,520	20,620	10,250	701,400	10,720	5,452	704	393	2,254	3,988	6,359
"02/11/07 CELLULOSE SY2H1"	75,080	31,360	24,930	375,200	2,948	1,459	649	400	2,095	7,150	8,334
"02/11/07 CELLULOSE SY2Z2"	73,650	28,060	22,240	337,700	3,598	1,066	714	426	1,663	5,975	5,195
"02/11/07 CELLULOSE BLOOD1"	128,300	29,240	4,941	2,803,000	6,377	15,490	688	447	735	3,213	10,030
"02/11/07 CELLULOSE BLOOD2"	101,900	26,030	5,736	2,218,000	6,518	7,604	714	468	817	4,711	2,713
"02/11/07 CELLULOSE GLSSTD1"	233,300	644,400	175,200	227,800	50,490	52,420	25,230	918	91,410	245,700	37,890
"02/11/07 CELLULOSE AIRBL3"	33,650	12,570	2,563	27,070	2,638	339	747	462	145	46	73
"02/11/07 CELLULOSE AIRBL4"	35,000	12,880	2,645	28,020	2,765	362	788	511	148	42	66
DRY											
"02/11/07 CELLULOSE AIRBL5"	25,660	10,520	2,381	23,830	2,197	327	860	511	145	95	74
"02/11/07 CELLULOSE AIRBL6"	26,490	10,700	2,466	24,380	2,211	338	831	532	128	41	73
"02/11/07 CELLULOSE BLANK5"	35,730	18,150	4,002	71,500	2,491	5,882	813	379	364	2,751	2,758
"02/11/07 CELLULOSE BLANK6"	38,820	18,460	4,104	76,720	2,500	5,450	882	366	348	2,147	2,319
"02/11/07 CELLULOSE SY2J3"	102,100	30,740	36,790	678,500	3,000	6,896	865	395	2,332	11,880	7,340
"02/11/07 CELLULOSE SY2J4"	117,400	36,750	43,590	791,600	3,104	5,782	948	466	2,869	14,010	8,050
"02/11/07 CELLULOSE BLOOD3"	107,400	32,000	4,320	2,893,000	6,533	8,471	929	539	382	1,056	3,126
"02/11/07 CELLULOSE BLOOD4"	106,200	33,000	4,300	2,766,000	6,308	7,468	967	540	392	1,173	3,369
"02/11/07 CELLULOSE GLSSTD7"	145,100	571,300	188,600	212,500	41,860	35,320	25,530	927	102,000	298,800	61,500
"02/11/07 CELLULOSE AIRBL7"	28,040	12,350	2,968	30,210	2,224	350	962	506	172	39	79
"02/11/07 CELLULOSE AIRBL8"	28,620	12,380	2,962	30,540	2,255	364	971	556	162	33	70
Ave SY2	71,975	14,940	36,137	680,940	557	673	59	62	2,246	10,496	5,157
Ave Blood	69,025	14,196	257	2,757,880	3,925	2,303	96	172	37	-1,332	709
Blank connected											
"02/11/07 CELLULOSE SY2J3"	64,325	12,435	32,737	604,390	505	1,230	17	27	1,977	9,431	4,802
"02/11/07 CELLULOSE SY2J4"	79,625	17,445	39,537	717,490	609	116	100	97	2,514	11,561	5,512
% Std Dev	15	24	13	12	13	117	100	79	17	14	10
"02/11/07 CELLULOSE BLOOD3"	69,625	13,895	267	2,823,880	4,038	2,805	81	171	37	-1,393	588
"02/11/07 CELLULOSE BLOOD4"	68,425	14,895	247	2,691,880	3,813	1,800	110	173	37	-1,270	831
% Std Dev	1	5	6	3	4	31	21	1	0	-7	24

Experiment 5A 1/1

APPENDIX EXPERIMENT 5B

Isotope - Raw Counts	Mg 24	Ca 44	Mn 55	Fe 56	Cu 66	Zn 66	As 76	Se 77	Mo 98	Ba 138	Pb 208
"02/11/07 CELLULOSE AIRBL5"	25,660	10,520	2,391	23,630	2,197	327	860	511	145	95	74
"02/11/07 CELLULOSE AIRBL6"	28,490	10,700	2,465	24,380	2,211	338	831	532	128	41	73
"02/11/07 CELLULOSE AIRBL5"	25,660	10,520	2,391	23,630	2,197	327	860	511	145	95	74
"02/11/07 CELLULOSE AIRBL6"	26,480	10,700	2,465	24,380	2,211	338	831	532	128	41	73
"02/11/07 CELLULOSE BLANK5"	35,730	18,150	4,002	71,500	2,491	5,882	813	378	364	2,751	2,758
"02/11/07 CELLULOSE BLANK6"	39,820	18,460	4,104	78,720	2,500	5,450	862	358	346	2,147	2,318
"02/11/07 CELLULOSE SY23"	102,100	30,740	36,790	678,900	3,000	6,096	948	465	2,332	11,080	7,340
"02/11/07 CELLULOSE SY24"	117,400	35,750	43,590	781,500	3,104	5,782	948	465	2,332	11,080	7,340
"02/11/07 CELLULOSE BLOOD3"	107,400	32,000	4,320	2,898,000	6,533	8,471	928	539	392	1,058	8,050
"02/11/07 CELLULOSE BLOOD4"	105,200	33,000	4,300	2,898,000	6,533	8,471	928	539	392	1,058	8,050
"02/11/07 CELLULOSE GLSST02"	145,100	571,300	166,600	212,500	41,650	35,320	25,530	827	102,000	288,800	81,500
"02/11/07 CELLULOSE AIRBL7"	28,040	12,350	2,966	30,210	2,224	350	962	505	172	39	79
"02/11/07 CELLULOSE AIRBL8"	28,620	12,380	2,982	30,640	2,255	364	971	556	162	33	70
Blank Corrected											
"02/11/07 CELLULOSE SY23"	64,326	12,435	32,737	604,380	505	1,230	17	27	1,977	9,431	4,802
"02/11/07 CELLULOSE SY24"	79,625	17,445	39,537	717,490	608	116	100	97	2,514	11,561	5,512
"02/11/07 CELLULOSE BLOOD3"	69,625	13,695	267	2,823,890	4,038	2,805	81	171	37	-1,393	588
"02/11/07 CELLULOSE BLOOD4"	68,425	14,695	247	2,691,890	3,813	1,800	110	173	37	-1,270	831
Conc in ppm in SY-2	2.69 (MgO) % in sample	7.06 (CaO)	0.32 (MnO)	2.43 (Fe2O3+FeO)	5.20	248.00	17.30	20.00	0.53	480.00	85.00
	0.60 % Metal in SY-2	0.71	0.77	0.70 0.78				conc ratio for SY-2		197.07	
Conc in ppm in SY-2	16220	56857	2478	17010 27689	5.20	248.00	17.30	20.00	0.53	480.00	85.00
Conc in ppm for SY-2 in 50mL sample	82.31	288.51	12.58	86.31 140.50	0.03	1.26	0.09	0.10	0.00	2.33	0.43
Average counts for SY-2	71975	14940	36137	660940	557	673	59	62	2246	10498	5157
Conc in ppm for blood samples (avg)	78.9	274	0.069	360	0.186	4.31	0.143	0.280	<0.001	<0.001	0.039
Expected concentrations for blood values where found in literature	50.0	320		500-1800	0.0-16	6.00					0.06

Experiment 5B/1

APPENDIX EXPERIMENT 12

Isotope - Raw Counts	Li 7	Mg 24	Ca 44	V 51	Cr 52	Mn 55	Fe 56	Cu 65	Zn 64	Ga 69	As 75	Sr 88	Zr 90	Mo 98	Cd 114
"02/11/27 HKH GLS STD 1"	107,400	194,900	680,900	182,200	152,900	252,900	258,100	41,720	25,830	183,900	25,180	415,400	177,500	112,700	36,070
"02/11/27 HKH GLS STD 2"	103,400	187,600	634,200	180,100	149,000	245,900	244,400	41,450	26,190	180,000	25,580	403,100	177,400	112,900	38,810
"02/11/27 HKH AIR BL 1"	1,919	84,140	21,220	122	1,638	10,620	50,120	1,434	1,245	231	3,055	1,761	139	252	188
"02/11/27 HKH AIR BL 2"	2,014	106,100	23,090	165	1,759	3,167	50,620	1,495	1,428	254	3,671	1,182	64	292	214
"02/11/27 HKH CELL ON BL 1"	2,024	101,800	27,540	235	6,289	1,602	61,880	1,602	1,984	445	2,785	1,161	241	341	4,647
"02/11/27 HKH CELL ON BL 2"	2,032	107,500	28,350	205	6,311	3,596	62,660	1,555	1,888	708	2,768	1,057	180	333	1,924
"02/11/27 HKH CELL ON BL 3"	1,986	82,580	24,860	233	5,007	2,827	54,740	1,353	1,381	235	3,257	1,026	87	288	455
"02/11/27 HKH CELL ON BL 4"	1,978	186,400	26,040	159	6,008	3,230	60,640	1,444	1,491	387	3,480	987	104	308	528
"02/11/27 HKH CELL ON BL 5"	2,213	118,000	37,410	1,080	7,191	4,522	79,450	1,482	1,778	568	3,531	1,489	100	325	1,183
"02/11/27 HKH CELL ON ME 1"	2,391	142,800	33,910	217	7,387	3,651	67,650	1,453	1,938	705	3,874	1,345	127	343	1,878
"02/11/27 HKH CELL ON ME 2"	4,348	122,000	33,410	4,217	10,980	5,811	52,480	1,988	2,422	2,658	3,073	7,990	3,179	2,037	1,690
"02/11/27 HKH CELL ON ME 3"	4,953	127,000	28,700	4,724	10,510	9,195	75,260	2,862	3,497	4,286	3,691	10,630	5,188	3,652	2,362
"02/11/27 HKH CELL ON ME 4"	4,805	130,600	30,080	5,087	11,280	10,120	68,430	2,768	3,823	4,783	4,319	13,940	5,819	3,817	2,714
"02/11/27 HKH CELL ON ME 5"	2,830	124,200	23,210	2,241	5,754	14,920	57,150	1,680	5,443	2,185	2,935	7,067	2,384	1,891	4,907
"02/11/27 HKH CELL ON ME 6"	3,703	131,200	33,780	3,760	10,320	13,870	73,610	4,235	6,735	3,644	4,100	8,289	4,292	2,345	4,865
"02/11/27 HKH GLS STD 3"	98,400	186,700	684,000	164,900	137,500	222,400	235,800	34,300	21,590	162,200	22,170	383,000	180,200	89,760	30,370
"02/11/27 HKH GLS STD 4"	92,880	188,600	646,500	177,600	147,600	243,100	237,900	39,800	26,360	192,200	25,820	442,700	192,600	114,900	38,260
"02/11/27 HKH AIR BL 3"	2,428	120,200	28,330	162	2,625	3,701	57,110	1,508	1,804	306	4,043	1,135	169	335	260
"02/11/27 HKH AIR BL 4"	2,051	123,100	24,890	184	3,245	3,691	57,590	1,503	1,749	302	3,952	6,418	88	376	238
Blank corrected															
"02/11/27 HKH CELL ON ME 1"	1,183	17,640	-6,755	2,531	301	2,032	-9,845	410	585	2,083	237	8,581	2,968	1,700	-1,588
"02/11/27 HKH CELL ON ME 2"	2,315	17,350	5,455	3,887	4,880	12,461	14,685	1,053	474	3,921	1,210	8,781	4,192	2,226	-1,851
"02/11/27 HKH CELL ON ME 3"	3,177	28,455	3,285	4,828	4,803	8,167	17,570	1,454	2,001	3,995	320	8,823	5,088	3,354	1,890
"02/11/27 HKH CELL ON ME 4"	3,019	30,055	4,635	4,881	6,673	7,082	11,740	1,390	2,487	4,472	951	12,333	5,719	3,519	2,222
"02/11/27 HKH CELL ON ME 5"	528	-6,700	-12,450	1,588	-1,535	10,884	-10,420	488	3,585	1,558	-668	5,645	2,251	1,357	3,312
"02/11/27 HKH CELL ON ME 6"	1,401	300	-1,880	3,107	3,031	9,784	60	2,763	4,877	3,007	408	8,867	4,179	2,011	3,330
Normalised to carbon															
"02/11/27 HKH CELL ON ME 1"	1,183	17,640	-6,755	2,531	301	2,032	-9,845	410	586	2,083	237	8,581	2,968	1,700	-1,588
"02/11/27 HKH CELL ON ME 2"	1,480	10,944	3,447	2,621	2,938	7,800	9,268	684	289	2,473	703	5,545	3,148	1,404	-1,188
"02/11/27 HKH CELL ON ME 3"	1,983	18,343	2,011	2,767	2,967	3,808	10,854	888	1,236	2,482	108	5,945	3,143	2,072	1,168
"02/11/27 HKH CELL ON ME 4"	1,722	17,144	2,644	2,700	3,179	4,045	6,867	793	1,419	2,551	642	7,035	3,262	2,007	1,268
"02/11/27 HKH CELL ON ME 5"	700	-8,880	-18,501	2,104	-2,034	14,358	-21,763	846	4,761	2,065	-805	7,402	2,883	1,788	4,468
"02/11/27 HKH CELL ON ME 6"	1,052	227	-1,425	2,355	2,258	7,418	45	2,095	3,606	2,280	377	5,207	3,188	1,525	2,524
Element - Raw Counts															
Li															
"02/11/27 HKH CELL ON ME 1"	1,279	22,329	-276,883	2,539	359	2,032	-10,736	1,330	2,100	3,465	237	7,987	5,775	7,065	-5,589
"02/11/27 HKH CELL ON ME 2"	1,678	13,853	165,727	2,529	3,088	7,880	10,108	2,155	1,072	4,115	783	8,713	6,127	5,824	-4,133
"02/11/27 HKH CELL ON ME 3"	2,122	20,885	88,675	2,808	3,808	9,808	11,837	2,815	4,431	4,056	189	7,197	6,115	8,580	4,089
"02/11/27 HKH CELL ON ME 4"	1,882	21,701	127,107	2,798	3,793	4,045	7,203	2,573	5,085	4,244	542	8,517	6,347	8,328	4,417
"02/11/27 HKH CELL ON ME 5"	757	-11,240	-783,309	2,111	-2,428	14,358	-23,732	2,098	17,090	3,437	-885	9,058	5,803	7,484	15,970
"02/11/27 HKH CELL ON ME 6"	1,148	289	-68,530	2,363	2,742	7,418	50	8,800	13,254	3,784	377	8,303	6,164	8,327	8,788
"02/11/27 HKH CELL ON ME 1"	1,279	22,329	-276,883	2,539	359	2,032	-10,736	1,330	2,100	3,465	237	7,987	5,775	7,065	-5,589

APPENDIX EXPERIMENT 12

Isotope - Raw Counts	Sn 120	Ba 138	La 139	Ce 140	Eu 161	Dy 162	Yb 174	Hf 178	Pb 206	U 238
¹⁰² 1127 HgH GLS STD 1"	182,100	399,900	450,200	517,100	270,700	112,100	128,100	91,780	64,500	115,800
¹⁰² 1127 HgH GLS STD 2"	188,400	398,000	439,100	507,500	263,900	105,600	123,400	88,580	65,130	119,100
¹⁰² 1127 HgH AIR BL 1"	141	1,144	36	13	18	13	9	4	312	21
¹⁰² 1127 HgH AIR BL 2"	152	163	25	20	20	9	14	14	28	8
¹⁰² 1127 HgH CELL ON BL 1"	675	1,180	182	164	112	45	53	32	4,450	96
¹⁰² 1127 HgH CELL ON BL 2"	665	1,673	142	138	52	21	23	24	4,769	83
¹⁰² 1127 HgH CELL R BL 1"	520	242	52	30	64	17	12	10	869	24
¹⁰² 1127 HgH CELL R BL 2"	508	264	44	26	38	14	11	33	771	18
¹⁰² 1127 HgH CELL UW BL 1"	355	635	58	83	50	24	29	14	2,590	45
¹⁰² 1127 HgH CELL UW BL 2"	474	947	63	118	163	22	14	10	2,789	187
¹⁰² 1127 HgH CELL ON ME 1"	3,088	6,293	7,892	7,442	4,328	1,952	2,202	1,708	4,944	1,805
¹⁰² 1127 HgH CELL ON ME 2"	4,887	9,724	12,560	11,710	6,768	3,268	3,531	2,848	5,061	2,346
¹⁰² 1127 HgH CELL R ME 1"	5,747	10,890	12,480	11,830	6,827	3,112	3,407	2,525	6,512	2,378
¹⁰² 1127 HgH CELL R ME 2"	8,991	11,620	13,930	12,810	7,818	3,587	3,897	2,858	6,501	2,716
¹⁰² 1127 HgH CELL UW ME 1"	3,485	5,400	5,898	5,602	3,258	1,482	1,577	1,167	9,840	1,200
¹⁰² 1127 HgH CELL UW ME 2"	5,174	10,489	9,953	9,717	5,474	2,845	2,812	2,111	7,553	1,833
¹⁰² 1127 HgH GLS STD 3"	180,000	374,500	437,100	473,100	258,400	105,700	118,500	85,230	47,700	68,150
¹⁰² 1127 HgH GLS STD 4"	203,100	433,000	497,200	557,500	295,800	120,200	138,200	100,200	64,190	123,100
¹⁰² 1127 HgH AIR BL 3"	718	287	41	22	34	18	9	10	44	9
¹⁰² 1127 HgH AIR BL 4"	738	463	98	17	32	13	10	12	833	8
Blank connected										
¹⁰² 1127 HgH CELL ON ME 1"	2,468	4,877	7,830	7,291	4,244	1,919	2,164	1,880	340	1,546
¹⁰² 1127 HgH CELL ON ME 2"	4,277	8,308	12,388	11,539	6,708	3,238	3,493	2,818	457	2,287
¹⁰² 1127 HgH CELL R ME 1"	5,228	10,757	12,432	11,802	6,778	3,067	3,398	2,503	5,882	2,366
¹⁰² 1127 HgH CELL R ME 2"	8,473	11,567	13,882	12,782	7,568	3,582	3,878	2,837	5,981	2,696
¹⁰² 1127 HgH CELL UW ME 1"	3,081	4,812	5,941	5,501	3,152	1,489	1,556	1,155	7,168	1,034
¹⁰² 1127 HgH CELL UW ME 2"	4,760	9,889	9,940	9,818	5,388	2,622	2,791	2,088	4,879	1,727
Normalized to cerium										
¹⁰² 1127 HgH CELL ON ME 1"	2,468	4,877	7,830	7,291	4,244	1,919	2,164	1,880	340	1,546
¹⁰² 1127 HgH CELL ON ME 2"	2,998	5,240	7,820	7,291	4,230	2,041	2,203	1,851	288	1,442
¹⁰² 1127 HgH CELL R ME 1"	3,230	6,533	7,880	7,291	4,188	1,913	2,098	1,547	3,516	1,455
¹⁰² 1127 HgH CELL R ME 2"	3,892	6,596	7,918	7,291	4,317	2,028	2,211	1,818	3,240	1,538
¹⁰² 1127 HgH CELL UW ME 1"	4,083	8,115	7,874	7,291	4,177	1,947	2,062	1,531	9,497	1,450
¹⁰² 1127 HgH CELL UW ME 2"	3,809	7,354	7,538	7,291	4,070	1,888	2,118	1,592	3,689	1,308
Element - Raw Counts										
¹⁰² 1127 HgH CELL ON ME 1"	7,572	9,801	7,838	8,238	8,879	7,525	8,804	6,154	848	1,566
¹⁰² 1127 HgH CELL ON ME 2"	8,278	7,508	7,828	8,238	8,850	8,004	8,804	8,049	560	1,452
¹⁰² 1127 HgH CELL R ME 1"	9,808	9,251	7,868	8,238	8,157	7,502	8,596	5,685	6,711	1,488
¹⁰² 1127 HgH CELL R ME 2"	11,328	9,202	7,908	8,238	9,031	7,944	9,802	5,928	9,184	1,549
¹⁰² 1127 HgH CELL UW ME 1"	12,524	8,528	7,881	8,238	8,739	7,534	8,484	5,608	18,124	1,460
¹⁰² 1127 HgH CELL UW ME 2"	11,070	10,257	7,544	8,238	8,514	7,798	8,654	5,530	7,089	1,318
¹⁰² 1127 HgH CELL ON ME 1"	7,572	9,801	7,838	8,238	8,879	7,525	8,804	6,154	848	1,566

APPENDIX EXPERIMENT 12

Isotope - Raw Counts	Li 7	Hg 204	Ca 44	V 51	Cr 52	Mn 55	Fe 56	Cu 65	Zn 66	Ga 69	As 76	Sr 88	Zr 90	Mo 98	Cd 114
"02/11/27 HKH CELL ON ME 2"	1,579	15,853	185,727	2,529	3,503	7,890	10,108	2,168	1,072	4,115	783	6,713	6,127	5,824	-4,133
Std dev	212	5,994	312,831	7	2,228	4,121	14,739	584	727	459	372	887	248	870	1,009
% Std dev.	15	33	-844	0	115	53	-4,633	34	44	12	74	12	4	14	-21
"02/11/27 HKH CELL R ME 1"	2,122	20,688	98,675	2,806	3,540	3,809	11,837	2,815	4,431	4,090	188	2,187	6,115	8,598	4,069
"02/11/27 HKH CELL R ME 2"	1,862	21,701	127,107	2,788	3,793	4,045	7,303	2,573	5,085	4,244	542	8,517	8,347	8,329	4,417
Std dev	184	716	21,519	5	179	167	3,208	242	462	105	242	833	164	180	248
% Std dev.	9	3	19	0	5	4	34	9	10	3	65	12	3	2	8
"02/11/27 HKH CELL UW ME 1"	757	-11,240	-703,309	2,111	-2,428	14,358	-23,732	2,098	17,030	3,437	-885	9,058	5,803	7,484	15,570
"02/11/27 HKH CELL UW ME 2"	1,148	288	-68,530	2,363	2,742	7,418	50	6,800	13,254	3,784	377	8,303	8,104	8,327	8,788
Std dev	277	8,152	512,498	178	3,655	4,908	16,816	3,325	2,870	253	682	1,948	255	804	4,780
% Std dev.	29	-148	-119	8	2,324	45	-142	76	18	7	-362	25	4	12	39

APPENDIX EXPERIMENT 12

Isotope - Raw Counts	Sr 120	Ba 138	La 139	Ce 140	Eu 151	Dy 162	Yb 174	Hf 178	Pb 208	U 238
¹⁰² 1127 HKH CELL ON ME 2"	8,276	7,308	7,828	8,238	8,660	8,004	8,828	8,048	550	1,452
Std dev	488	369	7	0	21	339	88	74	70	74
% Std dev	6	5	0	0	0	4	1	1	12	5
¹⁰² 1127 HKH CELL R ME 1"	9,909	9,251	7,688	8,238	8,757	7,502	6,596	5,885	8,711	1,488
¹⁰² 1127 HKH CELL R ME 2"	11,328	9,202	7,828	8,238	8,031	7,944	6,932	5,828	6,184	1,548
Std dev	1,002	35	169	0	194	313	251	183	372	59
% Std dev	9	0	2	0	2	4	4	3	6	4
¹⁰² 1127 HKH CELL UW ME 1"	12,524	8,526	7,881	8,238	8,738	7,634	6,484	5,809	18,124	1,480
¹⁰² 1127 HKH CELL UW ME 2"	11,070	10,257	7,544	8,238	8,514	7,788	6,854	5,830	7,059	1,319
Std dev	1,009	1,224	239	0	159	114	120	157	7,824	100
% Std dev	9	13	3	0	2	1	2	3	62	7

APPENDIX EXPERIMENT 16A

UNWASHED MATRICES AR and NH4F Beaks																									
Element	Run Counts	Li	Mg	Ca	V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Sr	Zr										
Class Standard																									
"02/12/09 HGH GLS STD 5"		189,379	178,885	54,275,269	282,339	275,238	382,091	373,770	400,083	202,827	157,619	34,725	28,845	648,428	423,431										
"02/12/09 HGH GLS STD 6"		213,282	186,388	56,148,258	288,275	283,518	380,856	390,116	408,883	221,517	131,886	36,200	28,882	688,743	440,172										
Air Blank																									
"02/12/09 HGH AIR BL 5"		5,181	25,573	1,614,554	142	3,237	3,867	40,942	238,677	10,401	1,904	1,528	21,405	651	387										
"02/12/09 HGH AIR BL 6"		5,671	28,489	1,730,516	147	3,348	4,246	42,909	271,177	10,806	1,824	1,583	22,334	739	363										
UW Blank																									
"02/12/09 HGH 3:1UW BL 1"		6,410	65,725	1,901,033	688	9,175	5,856	252,286	280,505	14,520	5,483	1,687	23,820	3,583	9,488										
"02/12/09 HGH 3:1UW BL 2"		6,882	67,228	1,920,188	725	7,705	5,676	241,388	284,748	14,208	5,764	1,915	23,867	3,245	10,630										
"02/12/09 HGH 3:1UW BL 3"		6,743	71,768	1,907,465	677	8,108	5,882	214,052	284,862	13,886	5,828	1,910	24,723	2,848	10,694										
% Std Dev		4	5	1	4	10	2	8	1	3	2	1	2	10	7										
UW AR W Blank																									
"02/12/09 HGH 3:1UW AR W BL 1"		5,226	34,313	1,513,148	1,273	6,831	4,885	228,219	258,142	10,877	3,317	1,819	19,707	3,838	10,498										
"02/12/09 HGH 3:1UW AR W BL 2"		5,477	38,142	1,589,014	1,383	7,821	5,461	249,889	275,535	11,918	3,242	1,864	18,477	3,678	11,138										
"02/12/09 HGH 3:1UW AR W BL 3"		5,181	34,517	1,534,742	1,340	7,288	5,448	240,581	282,780	11,828	3,445	1,873	19,568	3,679	12,919										
% Std Dev		3	6	2	4	7	5	5	5	4	3	2	1	2	7										
UW NH4F W Blank																									
"02/12/09 HGH 3:1UW NH4F W BL 1"		4,803	36,102	1,557,277	1,087	7,811	5,174	141,976	263,991	10,428	3,580	1,754	20,657	3,485	9,284										
"02/12/09 HGH 3:1UW NH4F W BL 2"		4,973	38,117	1,583,756	1,123	7,268	5,620	151,887	286,088	11,956	3,689	1,839	20,088	3,628	10,510										
"02/12/09 HGH 3:1UW NH4F W BL 3"		4,881	38,089	1,547,887	1,141	7,817	5,781	157,858	278,838	10,110	3,448	1,861	19,109	3,411	9,505										
% Std Dev		2	3	1	2	4	6	6	5	3	6	6	4	3	7										
UW ME 1ppm																									
"02/12/09 HGH 3:1UW ME 1"		7,561	72,125	1,911,268	6,278	14,004	11,730	255,463	288,047	15,623	6,448	3,265	25,438	19,822	20,543										
"02/12/09 HGH 3:1UW ME 2"		7,351	77,252	1,946,540	6,859	14,315	12,580	266,171	305,178	16,114	6,203	3,201	25,868	21,572	25,118										
"02/12/09 HGH 3:1UW ME 3"		7,388	78,018	1,980,141	5,947	14,589	11,290	324,956	284,918	15,455	6,483	4,163	28,758	17,926	19,321										
% Std Dev		1	4	1	6	2	6	13	13	2	2	14	3	9	14										
UW AR W ME 1ppm																									
"02/12/09 HGH 3:1UW AR W ME 1"		5,988	38,677	1,812,723	4,321	8,712	8,003	310,424	286,069	12,382	4,775	2,840	20,254	11,389	18,218										
"02/12/09 HGH 3:1UW AR W ME 2"		5,757	40,359	1,828,377	4,503	8,887	8,084	285,127	285,876	12,336	4,581	2,864	20,038	11,668	18,468										
"02/12/09 HGH 3:1UW AR W ME 3"		5,618	41,374	1,869,659	4,047	8,887	8,185	283,238	288,857	11,948	4,228	2,742	20,428	10,992	18,098										
% Std Dev		2	3	1	5	1	1	5	5	1	6	2	1	3	3										
UW NH4F W ME 1ppm																									
"02/12/09 HGH 3:1UW NH4F W ME 1"		5,522	48,185	1,617,840	2,446	8,874	6,383	188,248	279,801	11,803	4,348	2,328	21,967	11,736	20,768										
"02/12/09 HGH 3:1UW NH4F W ME 2"		5,772	47,201	1,623,005	2,757	8,578	6,321	177,228	278,894	11,391	4,624	2,952	21,059	11,544	19,966										
"02/12/09 HGH 3:1UW NH4F W ME 3"		5,740	47,048	1,604,225	2,815	8,882	6,131	173,531	283,830	11,456	4,478	2,435	22,157	11,365	20,151										
% Std Dev		2	1	1	6	2	2	5	5	1	3	2	3	2	2										
Matrix corrected																									
UW ME minus Av. UW Blank																									

Experiment 16A/1

APPENDIX EXPERIMENT 16A

"URWASHED" MATRICES AR and NH4F Base																											
Element - Raw Counts																											
Glass Standard																											
"021209 HKH GLS STD 5"																											
558,331	149,498	551,101	550,080	434,698	560,157	480,267	321,167	280,733	205,884	352	56,336	51,285															
563,278	154,043	559,025	565,925	442,521	565,630	487,238	333,957	272,971	213,268	349	56,243	55,019															
Air Blank																											
1,533	248	654	221	60	31	32	66	31	58																		
1,733	187	590	188	50	33	39	38	44	28																		
UW Blank																											
2,971	1,271	7,988	6,275	335	1,223	60	43	56	737	734	3,078	372															
2,871	750	8,285	11,414	232	384	69	41	52	941	734	1,578	285															
2,945	716	6,930	15,280	221	307	45	63	42	532	778	908	218															
2	34	10	41	24	80	21	24	14	28	3	58	26															
UW AR W Blank																											
2,989	483	10,783	1,589	189	172	66	51	25	557	552	637	428															
3,288	330	11,289	1,040	74	139	37	48	33	575	532	488	508															
3,355	396	11,550	1,153	74	112	36	24	41	604	627	584	546															
6	16	3	23	39	21	36	36	24	4	9	13	12															
UW NH4F W Blank																											
2,923	304	7,988	1,834	219	121	40	19	39	577	604	453	407															
3,128	307	8,341	2,171	167	140	38	34	52	589	845	542	434															
3,419	287	23,091	1,868	165	223	51	58	21	548	855	478	380															
8	6	67	9	17	33	18	53	41	2	19	8	5															
UW ME 1ppm																											
22,088	7,178	28,544	24,883	21,149	25,204	24,639	21,167	18,427	19,288	1,088	4,458	3,335															
21,542	6,185	27,031	28,278	20,588	24,311	25,309	21,242	18,181	18,983	958	5,824	3,288															
23,382	6,368	30,788	25,370	20,288	25,615	21,624	17,618	15,157	17,404	1,177	6,019	3,041															
4	6	5	3	2	3	8	10	11	6	8	15	5															
UW AR W ME 1ppm																											
21,554	5,139	26,715	13,515	10,910	12,185	11,555	9,432	8,103	8,549	1,482	3,983	2,082															
22,767	5,950	26,257	14,683	10,570	12,818	12,813	10,239	8,888	8,711	1,560	4,049	2,220															
21,928	6,096	26,152	13,977	10,746	12,381	12,218	9,253	8,028	8,675	1,488	4,149	2,200															
3	9	1	4	2	3	5	6	6	1	3	2	3															
UW NH4F W ME 1ppm																											
11,983	3,488	16,239	13,505	7,970	14,278	13,789	11,148	9,190	8,619	1,014	2,928	1,566															
11,567	3,074	13,863	14,725	8,941	14,843	13,257	10,159	8,944	8,481	1,051	3,085	1,714															
11,896	3,315	16,285	13,719	9,101	14,354	13,612	11,101	9,198	8,769	1,043	3,274	1,628															
2	7	1	5	7	2	2	5	5	6	2	6	6															
Matrix corrected UW ME minus Ar, UW Blank																											

APPENDIX EXPERIMENT 16A

Element - Raw Counts	Li	Mg	Na	Ca	V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Sr	Zr
UW ME1	879	3,885	1,706	5,566	5,341	5,892	19,559	12,676	1,482	823	1,302	1,468	1,302	16,364	10,272
UW ME2	679	9,012	35,978	8,019	5,652	6,742	30,207	21,804	1,973	578	1,304	1,831	1,831	18,314	14,845
UW ME3	717	7,778	18,421	5,257	5,926	5,452	88,052	11,544	1,313	858	2,266	2,266	2,266	14,869	9,050
% Std Dev	14	39	459	7	5	11	81	37	22	20	31	35	35	11	27
UW AR W ME minus UW AR W Blank															
UW AR W ME1	695	3,020	73,756	2,998	1,259	2,791	71,536	13,823	888	1,440	888	673	673	7,657	7,999
UW AR W ME2	456	4,712	90,610	3,177	1,555	2,762	59,238	13,730	886	1,248	1,012	457	457	7,936	7,249
UW AR W ME3	517	5,717	70,882	2,722	1,534	2,883	44,350	16,512	475	894	880	847	847	7,260	6,878
% Std Dev	21	30	14	8	7	2	24	11	31	23	7	30	30	4	8
UW NHAF W ME minus UW NHAF W Blank															
UW NHAF W ME1	636	9,383	54,867	1,329	1,378	885	39,949	10,897	759	708	477	2,018	2,018	8,235	11,002
UW NHAF W ME2	896	10,428	60,031	1,940	1,260	783	28,629	10,768	647	986	501	1,108	1,108	8,043	10,229
UW NHAF W ME3	854	10,278	41,252	1,498	1,554	603	21,930	14,928	612	840	584	2,206	2,206	7,864	10,385
% Std Dev	17	6	19	10	10	18	30	19	17	16	11	33	33	2	4
Blank Corrected															
Normalized to Average Calcium															
UW ME1-UW BL1	873	3,859	1,695	5,550	5,308	5,853	19,431	12,593	1,472	818	1,458	1,283	1,283	18,257	10,205
UW ME2-UW BL2	700	9,291	37,091	6,195	5,827	6,951	31,203	22,478	2,834	596	1,344	1,888	1,888	18,881	13,305
UW ME3-UW BL3	701	7,800	18,978	5,197	5,791	5,327	87,013	11,280	1,293	839	2,214	2,559	2,559	14,333	9,943
% Std Dev	43	40	423	9	5	14	78	40	24	18	33	33	33	14	30
UW AR W ME1-UW AR W BL1	702	3,036	75,631	3,072	1,394	2,862	73,335	14,277	910	1,477	1,013	681	681	7,852	8,203
UW AR W ME2-UW AR W BL2	441	4,558	87,854	3,074	1,484	2,872	54,404	13,282	837	1,205	979	442	442	7,877	7,613
UW AR W ME3-UW AR W BL3	522	5,768	71,530	2,746	1,548	2,888	44,720	18,860	479	902	868	856	856	7,326	6,939
% Std Dev	29	36	11	6	5	4	26	12	31	24	6	31	31	4	10
UW NHAF W ME1-UW NHAF W BL1	648	9,535	55,697	1,349	1,397	868	39,233	11,062	770	719	484	2,047	2,047	8,360	11,765
UW NHAF W ME2-UW NHAF W BL2	865	10,176	58,584	1,801	1,249	774	25,991	10,502	534	983	488	1,032	1,032	7,850	9,984
UW NHAF W ME3-UW NHAF W BL3	862	10,375	41,653	1,513	1,569	809	22,143	15,071	618	849	589	2,227	2,227	7,940	10,486
% Std Dev	16	4	17	9	11	17	31	20	19	14	11	35	35	3	6
Percent Standard Deviations															
Matrix Blank															
Av. UW BL %STDEV	4	6	1	4	10	2	8	1	3	2	1	2	2	10	7
Av. UW AR WASH BL %STDEV	3	6	2	4	7	6	5	5	4	3	2	1	1	2	7
Av. UW NHAF WASH BL %STDEV	2	3	1	2	4	6	5	3	9	6	6	4	4	3	7
1 ppm Matrix element Standard															
Av. UW ME %STDEV	1	4	1	6	2	6	13	2	2	2	14	3	3	9	14
Av. UW AR W ME %STDEV	2	3	1	5	1	1	5	1	2	6	2	2	2	3	3
Av. UW NHAF W ME %STDEV	2	1	1	6	2	2	5	1	1	3	2	2	2	2	2
Matrix Blank Corrected															
Av. UW ME-UW BL %STDEV	14	39	459	7	5	11	81	37	22	20	31	35	35	11	27
Av. UW AR W ME-UW AR W BL %STDEV	21	30	14	8	7	2	24	11	31	23	7	30	30	4	8
Av. UW NHAF W ME-UW NHAF W BL %STDEV	17	6	19	10	10	18	30	19	17	16	11	33	33	2	4

APPENDIX EXPERIMENT 16A

Element - Raw Counts	Mo	Cd	Sn	Ba	La	Ce	Eu	Dy	Yb	Hf	Hg	Pb	U
UW ME1	20,067	6,268	20,848	14,003	20,887	24,588	24,581	21,118	18,376	18,529	339	2,569	3,043
UW ME2	18,612	5,273	20,234	15,288	20,326	23,873	25,250	21,193	18,141	17,927	249	3,736	3,006
UW ME3	20,482	5,454	23,038	14,380	20,024	24,977	21,868	17,570	15,107	16,687	428	4,131	2,749
% Std Dev	5	9	7	5	2	3	8	10	11	5	26	23	5
UW AR W ME minus UW AR W Blank													
UW AR W ME1	18,343	4,740	15,508	12,236	10,814	12,064	11,509	9,391	8,070	7,970	912	3,413	1,599
UW AR W ME2	19,558	5,561	15,060	13,322	10,474	12,777	12,767	10,188	8,865	8,132	980	3,480	1,727
UW AR W ME3	18,718	4,885	15,444	12,716	10,650	12,250	12,189	9,212	7,996	8,086	888	3,578	1,707
% Std Dev	3	10	2	4	2	3	6	5	6	1	5	2	4
UW NH4F W ME minus UW NH4F W Blank													
UW NH4F W ME1	8,808	3,179	3,200	11,514	7,787	14,116	13,745	11,112	8,153	8,042	313	2,437	1,158
UW NH4F W ME2	8,410	2,705	2,824	12,734	8,757	14,981	13,214	10,122	8,907	8,903	350	2,584	1,304
UW NH4F W ME3	8,738	3,008	3,245	11,722	8,917	14,182	13,789	11,064	9,071	8,181	341	2,783	1,215
% Std Dev	2	8	7	5	7	2	2	5	5	7	5	7	6
Blank Corrected													
Normalised to Average Calcium													
UW ME1-UW BL1	19,035	6,225	20,710	13,912	20,750	24,405	24,420	20,880	18,256	18,408	337	2,552	3,024
UW ME2-UW BL2	19,188	5,438	20,880	15,761	20,866	24,405	24,032	21,846	18,702	18,481	256	3,851	3,088
UW ME3-UW BL3	19,884	5,329	22,566	14,051	19,566	24,405	21,073	17,167	14,781	16,285	418	4,036	2,888
% Std Dev	2	9	5	7	4	0	11	12	13	7	24	23	7
UW AR W ME1-UW AR W BL1	18,910	4,880	15,802	12,568	11,019	12,380	11,802	9,830	8,275	8,173	836	3,500	1,839
UW AR W ME2-UW AR W BL2	18,918	5,379	14,569	12,888	10,132	12,350	12,351	9,865	8,576	7,887	858	3,368	1,870
UW AR W ME3-UW AR W BL3	18,887	4,758	15,884	12,831	10,746	12,360	12,279	9,285	8,068	8,188	906	3,610	1,722
% Std Dev	6	7	6	1	5	0	8	3	3	2	3	4	2
UW NH4F W ME1-UW NH4F W BL1	8,809	3,227	3,248	11,688	7,804	14,330	13,853	11,280	8,282	8,163	317	2,474	1,173
UW NH4F W ME2-UW NH4F W BL2	8,208	2,640	2,756	12,428	8,548	14,330	12,698	9,880	8,870	8,880	341	2,532	1,273
UW NH4F W ME3-UW NH4F W BL3	8,824	3,035	3,277	11,838	8,004	14,330	13,802	11,171	8,159	8,281	345	2,810	1,227
% Std Dev	6	10	9	3	7	0	4	7	3	6	4	7	4
Percent Standard Deviations													
Matrix Blank													
Av. UW BL %STDDEV	2	34	10	41	24	80	21	24	14	28	3	58	28
Av. UW AR WASH BL %STDDEV	8	16	3	23	38	21	36	36	24	4	9	13	12
Av. UW NH4F WASH BL %STDDEV	8	8	87	9	17	33	16	53	41	2	19	8	5
1ppm Multi-element Standard													
Av. UW ME %STDDEV	4	8	5	3	2	3	8	10	11	5	8	15	5
Av. UW AR W ME %STDDEV	3	8	1	4	2	3	5	5	6	1	3	2	3
Av. UW NH4F W ME %STDDEV	2	7	1	5	7	2	2	5	5	8	2	6	5
Matrix Blank Corrected													
Av. UW ME-UW BL %STDDEV	5	9	7	5	2	3	8	10	11	5	26	23	5
Av. UW AR W ME-UW AR W BL %STDDEV	3	10	2	4	2	3	5	5	6	1	5	2	4
Av. UW NH4F W ME-UW NH4F W BL %STDDEV	2	8	7	5	7	2	2	5	5	7	6	7	6

APPENDIX EXPERIMENT 16A

Element - Raw Counts	Li	Mg	Ca	V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Sr	Zr
Matrix Blank Corrected														
Normalized to Average Carbon														
Ax. UW ME-JW BL %STDEV	13	40	428	9	5	14	79	40	24	18	28	33	14	30
Ax. UW AR W ME-UW AR W BL %STDEV	24	30	11	8	5	4	25	12	31	24	6	31	4	10
Ax. UW NHF ME-UW NHF W BL %STDEV	16	4	17	8	11	17	31	20	19	14	11	35	3	6

APPENDIX EXPERIMENT 16A

Element - Row Counts	No	Cd	Sn	Ba	La	Ce	Eu	Dy	Yb	Hf	Mg	Pb	U
Matrix Blank Corrected													
Normalised to Average Cerium													
Av. UW ME-UW BL %STDEV	2	8	5	7	4	0	11	12	13	7	24	23	7
Av. UW AR W ME-UW AR W BL %STDEV	0	7	5	1	5	0	2	3	3	2	3	4	2
Av. UW NH4F ME-UW NH4F W BL %STDEV	5	10	9	3	7	0	4	7	3	6	4	7	4

Experiment 16B.1

"WASHED" MATRICES															
AR and NH4F Base															
Element - Raw Counts															
Glass Standard															
	Li	Mg	Ca	V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Sr	Zr	Mo
"02121209 HCH GLS STD 1"	220,284	194,784	59,436,620	314,958	288,900	401,800	408,763	421,254	231,058	155,843	30,110	29,424	704,094	474,787	629,583
"02121209 HCH GLS STD 2"	195,177	172,010	51,381,502	268,845	263,491	344,900	358,392	390,073	197,024	128,515	33,110	29,310	618,338	392,164	515,417
"02121209 HCH GLS STD 3"	202,475	179,353	54,340,745	289,937	278,398	370,025	381,396	394,180	210,348	144,055	35,673	27,894	859,008	436,212	574,209
"02121209 HCH GLS STD 4"	190,128	174,342	52,500,302	272,984	282,040	350,007	358,280	388,740	198,445	130,148	33,004	27,778	616,183	400,778	528,898
Air Blank															
"02121209 HCH AIR BL 1"	5,977	18,471	2,848,357	202		5,461	48,121	247,806	11,395	1,831	1,864	25,281	613	507	1,482
"02121209 HCH AIR BL 2"	5,538	18,628	2,739,808	213	2,833	5,482	48,459	234,151	11,094	1,877	1,657	23,271	644	543	1,364
"02121209 HCH AIR BL 3"	5,764	18,270	2,817,840	184	3,170	5,065	48,307	238,876	11,478	1,827	1,586	25,527	643	468	1,417
"02121209 HCH AIR BL 4"	5,528	18,208	2,701,878	181	3,380	5,039	48,143	237,853	11,044	1,800	1,692	25,125	672	563	1,351
W Blank															
"02121209 HCH 3:1W BL 1"	8,847	31,351	3,534,742	859	11,022	9,277	372,705	202,256	13,280	7,105	1,477	23,621	2,430	7,098	3,486
"02121209 HCH 3:1W BL 2"	8,458	32,613	3,773,709	783	10,525	9,020	382,888	208,086	12,355	6,717	1,595	23,198	2,388	7,071	3,107
"02121209 HCH 3:1W BL 3"	8,980	34,293	2,878,343	714	9,206	8,719	243,007	213,852	12,850	6,283	1,649	21,771	2,058	6,880	3,432
% Std Dev	4	3	14	9	9	3	24	3	3	6	6	4	9	3	8
W AR W Blank															
"02121209 HCH 3:1W AR W BL 1"	6,247	28,697	3,495,305	635	11,745	8,721	637,549	193,118	11,822	8,802	1,828	22,404	6,382	14,024	2,279
"02121209 HCH 3:1W AR W BL 2"	6,575	28,763	3,237,089	888	11,349	8,574	674,148	201,982	11,990	8,040	1,834	21,178	5,254	15,559	2,083
"02121209 HCH 3:1W AR W BL 3"	8,282	28,738	3,140,376	872	10,750	8,780	687,041	228,748	12,098	8,018	1,817	22,803	5,604	18,370	2,338
% Std Dev	3	2	6	23	4	1	2	8	1	7	6	3	3	8	6
W NH4F W Blank															
"02121209 HCH 3:1W NH4F W BL 1"	5,772	32,181	2,687,136	715	10,309	7,167	437,587	197,859	11,220	4,750	1,504	22,482	3,654	11,498	2,201
"02121209 HCH 3:1W NH4F W BL 2"	6,388	31,984	2,355,399	784	9,820	6,783	423,514	212,844	11,330	5,498	1,620	23,528	3,228	9,050	1,956
"02121209 HCH 3:1W NH4F W BL 3"	5,754	33,033	2,640,378	749	10,777	7,180	441,980	220,031	11,744	4,808	1,702	23,905	3,487	10,522	1,888
% Std Dev	6	2	7	5	5	3	2	5	2	8	6	3	5	12	8
W ME 1 ppm															
"02121209 HCH 3:1W ME1"	7,407	33,210	3,415,862	618	11,687	10,350	423,285	224,732	14,384	7,091	3,857	23,948	11,848	28,487	15,350
"02121209 HCH 3:1W ME2"	7,317	35,078	3,384,507	626	11,531	10,100	403,051	232,219	15,283	7,527	3,040	23,659	12,112	24,263	14,388
"02121209 HCH 3:1W ME3"	7,156	30,751	3,530,986	639	11,309	10,870	413,820	231,651	14,552	6,948	3,273	24,078	11,879	28,687	13,708
% Std Dev	2	7	2	2	2	4	2	2	3	3	12	1	2	9	6
W AR W ME 1 ppm															
"02121209 HCH 3:1W AR W ME1"	6,887	30,218	3,165,728	923	11,553	9,738	700,616	215,198	13,858	8,783	3,855	22,198	10,713	21,451	17,728
"02121209 HCH 3:1W AR W ME2"	6,841	28,188	2,871,582	882	11,470	9,803	710,201	223,068	15,406	8,937	3,516	21,882	11,472	20,403	17,363
"02121209 HCH 3:1W AR W ME3"	6,770	29,931	3,155,888	921	11,885	10,125	707,124	228,478	14,008	7,057	4,058	21,870	12,085	22,401	17,808
% Std Dev	1	4	4	4	3	2	1	3	5	2	8	1	6	5	1
OW NH4F W ME 1 ppm															
"02121209 HCH 3:1W NH4F W ME1"	6,604	37,038	2,717,840	813	11,887	9,924	484,047	219,982	15,183	5,783				21,839	19,517
"02121209 HCH 3:1W NH4F W ME2"	6,541	40,140	2,814,554	759	11,391	10,510	472,312	222,394	14,869	5,430	2,889			22,148	18,588
"02121209 HCH 3:1W NH4F W ME3"	6,882	32,443	2,686,531	833	12,348	10,350	508,540	232,492	16,503	5,851	2,853			23,507	20,798
% Std Dev	3	11	2	5	4	3	4	3	6	5	7	1	3	4	5
Matrix corrected															

APPENDIX EXPERIMENT 16B

"WASHED" MATRICES													
AR and NH4F Balco													
Element - Row Counts													
Glass Standard													
	Cd	Sn	Ba	La	Ce	Eu	Dy	Yb	Hf	Hg	Pb	U	
"021209 HKH GLS STD 1"	170,782	618,441	602,149	470,824	624,435	515,177	392,123	291,677	229,159	314	61,832	56,235	
"021209 HKH GLS STD 2"	132,893	529,258	517,025	389,860	508,447	437,722	295,536	246,074	192,251	368	60,630	50,816	
"021209 HKH GLS STD 3"	158,447	551,530	585,007	439,868	582,401	482,705	334,323	274,448	214,616	348	58,138	54,868	
"021209 HKH GLS STD 4"	136,363	525,190	514,198	396,815	527,378	436,162	299,847	244,039	191,744	328	50,404	48,569	
AR Blank													
"021209 HKH AR BL 1"	272	538	249	60	44	48	63	30	35	282	96	9	
"021209 HKH AR BL 2"	220	542	241	53	36	63	36	26	32	224	81	13	
"021209 HKH AR BL 3"	222	529	178	45	27	39	55	27	28	233	84	8	
"021209 HKH AR BL 4"	244	537	183	58	28	39	55	37	34	303	97	0	
W Blank													
"021209 HKH 3:1W BL 1"	1,351	7,888	3,111	231	283	73	79	96	260	394	1,049	148	
"021209 HKH 3:1W BL 2"	1,243	8,119	3,205	199	282	71	84	54	295	394	1,182	182	
"021209 HKH 3:1W BL 3"	1,117	6,846	3,884	189	228	68	81	90	269	382	1,029	143	
% Std Dev	8	9	12	8	13	4	3	5	7	0	8	14	
W AR W Blank													
"021209 HKH 3:1W AR W BL 1"	2,183	15,584	1,924	74	189	88	80	35	577	448	2,268	804	
"021209 HKH 3:1W AR W BL 2"	1,887	15,098	1,868	82	214	84	53	44	530	460	1,885	677	
"021209 HKH 3:1W AR W BL 3"	1,807	16,187	2,094	88	224	54	60	48	538	408	1,800	705	
% Std Dev	10	4	13	7	8	10	7	15	4	6	12	9	
W NH4F W Blank													
"021209 HKH 3:1W NH4F W BL 1"	726	9,260	2,169	111	183	49	82	42	474	394	1,721	390	
"021209 HKH 3:1W NH4F W BL 2"	858	9,311	2,173	98	174	42	81	41	431	484	1,582	358	
"021209 HKH 3:1W NH4F W BL 3"	863	8,855	1,781	90	175	49	70	42	451	431	1,981	378	
% Std Dev	10	3	11	11	3	8	9	3	5	8	12	4	
W ME 1ppm													
"021209 HKH 3:1W ME 1"	5,072	19,911	12,882	8,703	12,082	10,890	8,885	7,352	7,558	781	3,457	1,897	
"021209 HKH 3:1W ME 2"	6,529	17,774	12,870	10,349	11,731	11,507	8,168	7,990	7,814	701	4,869	1,564	
"021209 HKH 3:1W ME 3"	4,088	17,828	12,706	10,088	11,888	10,888	8,433	7,217	7,802	683	2,809	1,526	
% Std Dev	23	7	1	3	2	3	4	5	3	10	28	5	
W AR W ME 1ppm													
"021209 HKH 3:1W AR W ME 1"	6,058	30,306	10,161	9,988	9,817	7,899	6,286	4,912	5,002	1,408	2,810	1,843	
"021209 HKH 3:1W AR W ME 2"	4,823	27,877	11,256	9,313	8,008	9,110	7,461	5,933	6,500	1,233	2,843	1,851	
"021209 HKH 3:1W AR W ME 3"	5,270	30,848	11,832	9,081	9,912	8,482	7,007	6,747	6,269	1,424	2,872	2,071	
% Std Dev	5	5	8	9	8	8	9	10	14	8	5	7	
W NH4F W ME 1ppm													
"021209 HKH 3:1W NH4F W ME 1"	5,817	37,458	17,885	13,202	18,704	15,018	11,821	9,988	8,840	1,068	3,038	2,888	
"021209 HKH 3:1W NH4F W ME 2"	6,528	40,952	17,948	14,303	17,689	15,556	11,708	9,984	9,645	969	3,725	2,417	
"021209 HKH 3:1W NH4F W ME 3"	5,770	35,951	16,827	13,882	18,984	15,867	12,168	9,993	8,030	1,065	3,305	2,713	
% Std Dev	7	6	3	4	3	3	2	1	4	8	10	6	
Matrix corrected													

Experiment 16B/2

APPENDIX EXPERIMENT 16B

Element - Raw Counts	Li	Mg	Ca	V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Sr	Zr	Mo
W ME minus Av. W Blank															
W ME1	641	-200	20,031	-171	1,415	1,325	67,082	16,841	1,276	390	2,287	1,102	8,053	21,537	12,003
W ME2	551	1,657	-11,424	-104	1,380	1,095	68,847	25,127	2,195	628	1,470	816	8,817	17,313	11,043
W ME3	390	-2,868	135,055	-150	1,050	1,865	77,117	23,560	1,464	248	1,703	1,235	9,384	21,747	10,384
% Std Dev	24	-537	161	-6	15	28	13	21	30	46	23	20	2	12	7
W AR W ME minus W AR W Blank															
W AR W ME1	355	817	-125,186	92	231	1,046	27,670	7,288	1,853	468	1,953	357	5,297	6,133	15,508
W AR W ME2	300	-1,232	-318,562	30	129	1,211	37,255	15,880	3,403	650	1,824	121	8,056	5,088	15,142
W AR W ME3	428	532	-135,065	90	713	1,433	34,178	20,829	2,905	771	2,386	109	8,658	7,083	15,388
% Std Dev	18	2,444	-57	49	57	16	15	48	28	24	14	68	11	16	1
W NHF W ME minus W NHF W Blank															
W NHF W ME1	629	4,643	163,537	64	1,585	2,887	49,887	9,748	3,752	774	801	796	10,736	11,449	17,508
W NHF W ME2	588	7,747	260,290	10	1,068	3,473	37,932	13,150	3,458	411	1,251	305	11,308	11,759	16,580
W NHF W ME3	887	50	145,227	84	2,048	3,353	74,180	22,248	5,072	803	1,355	869	11,548	13,117	18,789
% Std Dev	26	93	33	73	30	10	34	43	21	38	17	43	4	7	8
Blank Corrected															
Normalized to Average Cerium															
W ME1	827	-198	19,610	-167	1,386	1,297	85,259	16,201	1,248	381	2,238	1,079	9,449	21,084	11,753
W ME2	556	1,672	-11,525	-165	1,302	1,104	67,440	25,850	2,214	631	1,483	823	8,904	17,487	11,141
W ME3	395	-2,703	136,783	-152	1,071	1,888	76,717	23,883	1,482	249	1,725	1,251	9,504	22,027	10,487
% Std Dev	23	-637	162	-5	14	23	12	22	31	46	21	20	3	12	6
W AR W ME1	381	900	-139,025	101	255	1,154	30,508	8,035	2,043	514	2,164	372	5,838	6,782	17,098
W AR W ME2	288	-1,177	-303,459	29	123	1,158	35,811	14,889	3,253	821	1,743	118	5,788	4,881	14,474
W AR W ME3	408	508	-129,019	88	891	1,369	32,551	19,708	2,775	738	2,260	104	6,370	6,767	14,700
% Std Dev	18	1,432	-52	53	53	10	8	41	23	18	13	77	5	14	9
W NHF W ME1	645	4,760	167,875	66	1,635	2,960	50,844	9,994	3,847	794	885	818	11,008	11,738	17,952
W NHF W ME2	547	7,492	251,689	9	1,053	3,359	38,703	12,717	3,325	397	1,210	285	10,804	11,372	16,035
W NHF W ME3	895	51	146,596	85	2,065	3,385	74,879	22,457	5,120	941	1,347	675	11,657	13,240	18,988
% Std Dev	26	82	29	74	32	7	36	44	23	40	16	45	4	8	6
Percent Standard Deviations															
Matrix Blank															
Av. W BL %STDEV	4	3	14	9	9	3	24	3	3	6	6	4	9	3	6
Av. W AR W BL %STDEV	3	2	6	22	4	1	2	9	1	7	7	6	3	8	6
Av. W NHF W BL %STDEV	6	2	7	5	5	3	2	5	2	8	6	3	5	12	8
100% Matrix Element Standard															
Av. W ME %STDEV	2	7	2	2	2	4	2	2	3	3	12	1	2	9	6
Av. W AR W ME %STDEV	1	4	4	4	3	2	1	3	5	2	8	1	6	5	1
Av. W NHF W ME %STDEV	3	11	2	5	4	3	4	3	6	5	7	1	3	4	6
Matrix Blank Corrected															
Av. W ME-W BL %STDEV	24	-537	161	-6	15	28	13	21	30	46	23	20	2	12	7
Av. W AR W ME-W AR W BL %STDEV	18	2,444	-57	49	57	16	15	48	28	24	14	68	11	16	1

Experiment 16B/3

APPENDIX EXPERIMENT 16B

Element - Raw Counts	Cd	Sn	Ba	La	Ce	Eu	Dy	Yb	Hf	Hg	Pb	U
W ME minus Av W Blank												
W ME1	3,835	12,380	9,292	9,494	11,814	10,819	8,814	7,286	7,283	388	2,389	1,540
W ME2	5,292	10,163	8,471	10,140	11,464	11,437	9,034	7,933	7,939	308	3,811	1,407
W ME3	2,852	10,217	9,306	9,880	11,419	10,928	8,352	7,181	7,327	470	1,721	1,360
% Std Dev	31	11	1	3	2	3	4	6	3	21	41	8
W AR W ME minus W AR W Blank												
W AR W ME1	3,105	14,851	8,318	6,887	8,408	7,638	6,237	4,870	4,447	967	623	1,201
W AR W ME2	2,871	12,251	9,382	8,233	9,887	9,049	7,424	5,841	5,945	794	865	1,208
W AR W ME3	3,388	15,221	9,970	8,000	9,703	8,421	6,950	5,708	5,714	985	885	1,428
% Std Dev	8	11	9	9	8	5	9	10	15	12	18	10
W NH4F W ME minus W NH4F W Blank												
W NH4F W ME1	5,002	28,316	15,854	13,102	16,527	14,968	11,744	8,767	8,388	658	1,281	2,283
W NH4F W ME2	5,713	31,410	15,887	14,203	17,522	15,816	11,630	9,352	9,083	538	1,971	2,042
W NH4F W ME3	4,954	25,709	14,888	13,783	16,787	15,840	12,088	9,881	9,578	638	1,550	2,337
% Std Dev	8	8	3	4	3	3	2	1	4	10	22	7
Blank Corrected												
Normalised to Average Cerium												
W ME1	3,755	12,041	8,098	9,294	11,585	10,591	8,628	7,142	7,130	380	2,320	1,508
W ME2	5,339	10,253	9,656	10,230	11,565	11,038	9,185	8,004	7,707	310	3,845	1,420
W ME3	2,888	10,349	9,428	10,007	11,565	11,038	8,459	7,253	7,421	477	1,743	1,387
% Std Dev	31	8	3	5	0	4	4	6	4	21	41	4
W AR W ME1	3,423	18,185	9,172	7,593	9,270	8,421	6,878	5,389	4,902	1,068	688	1,324
W AR W ME2	2,744	11,711	8,978	7,869	9,270	8,850	7,086	5,678	5,683	758	818	1,156
W AR W ME3	3,218	14,540	9,624	7,843	9,270	8,045	6,839	5,451	5,458	941	845	1,385
% Std Dev	11	16	3	2	0	4	3	3	8	17	11	8
W NH4F W ME1	5,128	28,033	10,050	13,434	18,945	15,348	12,041	10,014	8,800	675	1,313	2,361
W NH4F W ME2	5,825	30,378	15,287	13,738	18,945	15,086	11,248	9,825	8,784	522	1,908	1,975
W NH4F W ME3	6,001	28,980	15,027	13,913	16,945	15,989	12,203	9,854	8,890	842	1,585	2,358
% Std Dev	5	6	3	2	0	3	4	2	1	13	19	10
Percent Standard Deviations												
Matrix Blank												
Av W BL %STDEV	8	9	12	8	13	4	3	5	7	0	8	14
Av W AR W BL %STDEV	10	4	13	7	8	10	7	15	4	6	12	9
Av W NH4F W BL %STDEV	10	3	11	11	3	8	9	3	5	8	12	4
Typical Multi-element Standard												
Av W ME %STDEV	23	7	1	3	2	3	4	5	3	10	20	6
Av W AR W ME %STDEV	5	5	8	8	8	8	8	10	14	6	5	7
Av W NH4F W ME %STDEV	7	6	3	4	3	3	2	1	4	6	10	6
Matrix Blank Corrected												
Av W ME-W BL %STDEV	31	11	1	3	2	3	4	6	3	21	41	8
Av W AR W ME-W BL %STDEV	8	11	9	9	8	8	9	10	15	12	18	10

APPENDIX EXPERIMENT 16B

Element - Raw Counts		Li	Mg	Ca	V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Br	Zr	Mo
Av. W NHAF ME-W NHAF W BL %STDEV		25	83	33	73	30	10	34	43	21	38	17	43	4	7	6
Matrix Blank Corrected																
Normalised to Average Cerium																
Av. W NE-W BL %STDEV		23	637	182	-5	14	29	12	22	31	40	21	20	3	12	6
Av. W AR W ME-W AR W BL %STDEV		18	1432	-52	53	83	10	8	41	23	18	13	71	5	18	9
Av. W NHAF ME-W NHAF W BL %STDEV		28	92	29	74	32	7	36	44	23	40	15	45	4	8	8

APPENDIX EXPERIMENT 16B

Element - Raw Counts	Cd	Sn	Ba	La	Ce	Eu	Dy	Yb	Hf	Hg	Pb	U
Av. W NHAF ME-W NHAF WBL %STDEV	8	8	3	4	3	3	2	1	4	10	22	7
Matrix Blank Connected												
Normalized to Average Count												
Av. W ME-WBL %STDEV	31	0	3	5	0	4	4	6	4	21	41	4
Av. W AR W ME-W AR WBL %STDEV	11	18	3	2	0	4	3	3	8	17	11	9
Av. W NHAF ME-W NHAF WBL %STDEV	5	6	3	2	0	3	4	2	1	13	19	10

APPENDIX EXPERIMENT 18

Isotope - Raw Counts	Li 7	Mg 24	Ca 44	V 51	Cr 52	Mn 55	Fe 56	Co 59	Ni 60	Cu 65	Zn 66
T0212/13 HKH GLS STD 1"	49,170	85,700	499,600	142,700	128,200	204,100	258,500	158,700	83,060	44,090	31,300
T0212/13 HKH AIR BL 1"	6,087	43,050	30,380	200	9,281	4,085	92,610	4,713	57,810	2,143	1,103
T0212/13 HKH AIR BL 2"	6,286	43,580	29,020	211	10,420	4,539	96,000	4,908	57,100	2,142	1,063
T0212/13 HKH BLOOD HEAT 1"	6,158	93,280	41,530	419	14,550	11,976	3,454,000	5,171	58,330	4,807	7,888
T0212/13 HKH BLOOD HEAT 2"	5,708	96,200	42,130	474	17,160	12,250	3,905,000	5,220	58,860	5,313	8,333
T0212/13 HKH BLOOD HEAT 3"	5,975	94,600	40,930	478	19,270	14,080	3,556,000	5,234	58,080	5,950	8,350
T0212/13 HKH BLOOD HEAT 4"	5,460	92,710	38,130	490	18,800	11,810	3,926,000	5,336	58,250	5,163	8,481
T0212/13 HKH BLOOD HEAT 5"	5,811	98,090	41,370	508	17,030	9,439	3,884,000	5,374	58,300	4,306	9,230
T0212/13 HKH BLOOD AIR 1"	5,142	104,600	43,810	475	19,060	11,200	3,502,000	5,280	59,010	5,641	9,320
T0212/13 HKH BLOOD AIR 2"	5,101	100,500	38,050	502	14,740	8,533	3,991,000	5,313	59,220	4,264	8,920
T0212/13 HKH BLOOD AIR 3"	5,364	124,400	40,090	460	16,840	9,338	3,497,000	5,382	59,480	4,139	9,310
T0212/13 HKH BLOOD AIR 4"	5,342	108,700	38,770	551	18,900	9,867	4,211,000	5,224	58,250	5,377	9,181
T0212/13 HKH BLOOD AIR 5"	5,469	111,100	38,580	628	18,710	9,405	4,763,000	5,337	59,630	4,642	8,860
T0212/13 HKH MATRIX BL"	4,983	36,400	31,890	713	13,480	9,868	477,300	4,168	57,880	2,435	1,796
T0212/13 HKH BLOOD 1" no matrix	5,276	102,900	39,780	245	13,780	5,998	2,779,000	4,441	58,110	5,066	8,127
T0212/13 HKH BLOOD 2" no matrix	5,511	133,500	52,230	267	14,890	6,401	3,997,000	4,568	58,050	7,003	12,500
T0212/13 HKH AIR BL 3"	5,574	37,660	23,580	280	12,450	6,069	110,100	4,932	57,120	1,930	1,602
T0212/13 HKH AIR BL 4"	5,882	38,930	24,410	268	12,770	6,228	111,000	5,120	57,100	1,980	1,653
T0212/13 HKH GLS STD 2"	42,650	66,880	435,700	122,900	108,000	176,500	236,400	128,300	78,170	37,790	24,760
Air Blank corrected											
T0212/13 HKH BLOOD HEAT 1"	169	52,290	14,815	179	3,115	6,872	3,350,950	251	1,220	2,746	8,536
T0212/13 HKH BLOOD HEAT 2"	-282	55,210	15,415	234	5,725	6,946	3,801,950	300	1,750	3,252	6,981
T0212/13 HKH BLOOD HEAT 3"	-15	53,810	14,215	238	7,835	8,776	3,452,950	314	970	3,889	6,988
T0212/13 HKH BLOOD HEAT 4"	-530	51,720	11,415	250	7,365	6,506	3,822,950	416	1,140	3,102	7,129
T0212/13 HKH BLOOD HEAT 5"	-379	57,090	14,635	266	5,595	4,135	3,790,950	454	1,190	2,245	7,878
T0212/13 HKH BLOOD AIR 1"	-848	63,610	17,095	236	7,625	6,898	3,398,950	360	1,900	3,580	7,968
T0212/13 HKH BLOOD AIR 2"	-898	58,510	11,335	263	3,305	3,229	3,887,950	393	2,110	2,203	7,568
T0212/13 HKH BLOOD AIR 3"	-628	83,410	13,375	220	5,405	3,034	3,393,950	442	2,370	2,078	7,958
T0212/13 HKH BLOOD AIR 4"	-648	67,710	12,055	311	7,465	4,363	4,107,950	304	1,150	3,316	7,829
T0212/13 HKH BLOOD AIR 5"	-621	70,110	11,865	388	7,275	4,101	4,659,950	417	2,520	2,581	7,598
Normalized to Ba											

APPENDIX EXPERIMENT 18

Isotope - Raw Counts	As 75	Se 78	Mo 98	Cd 114	Sn 120	Sb 121	Ba 138	La 139	Ce 140	Eu 151	Dy 162
¹⁰² 12/13 HKH GLS STD 1"	99,680	11,340	132,300	69,000	214,900	200,200	438,300	500,900	551,600	258,000	88,710
¹⁰² 12/13 HKH AIR BL 1"	4,160	12,590	813	517	750	92	167	88	60	28	14
¹⁰² 12/13 HKH AIR BL 2"	4,254	12,580	868	538	649	91	163	108	85	33	21
¹⁰² 12/13 HKH BLOOD HEAT 1"	15,560	13,380	1,520	644	2,127	321	821	228	210	41	18
¹⁰² 12/13 HKH BLOOD HEAT 2"	16,640	13,790	2,005	631	2,142	407	964	222	144	37	10
¹⁰² 12/13 HKH BLOOD HEAT 3"	41,150	13,920	1,801	571	2,202	261	938	217	259	31	13
¹⁰² 12/13 HKH BLOOD HEAT 4"	22,330	13,960	2,050	561	1,915	217	914	145	109	31	22
¹⁰² 12/13 HKH BLOOD HEAT 5"	20,760	14,380	2,160	684	2,051	341	853	162	129	47	12
¹⁰² 12/13 HKH BLOOD AIR 1"	19,110	13,590	1,624	641	2,201	261	876	176	119	45	16
¹⁰² 12/13 HKH BLOOD AIR 2"	19,860	13,770	1,484	618	2,032	338	808	168	157	34	14
¹⁰² 12/13 HKH BLOOD AIR 3"	29,070	14,830	1,589	614	2,003	448	874	170	173	46	18
¹⁰² 12/13 HKH BLOOD AIR 4"	27,000	14,470	1,655	673	2,381	335	888	242	256	37	17
¹⁰² 12/13 HKH BLOOD AIR 5"	24,150	14,730	1,854	672	2,290	227	939	178	179	39	25
¹⁰² 12/13 HKH MATRIX BL"	30,810	13,060	2,809	640	3,371	251	504	160	133	71	17
¹⁰² 12/13 HKH BLOOD 1" no matrix	12,770	8,767	998	752	974	270	1,672	180	74	32	18
¹⁰² 12/13 HKH BLOOD 2" no matrix	18,230	11,140	1,138	725	1,268	283	2,175	214	82	34	20
¹⁰² 12/13 HKH AIR BL 3"	5,313	12,780	902	540	725	79	191	130	69	30	18
¹⁰² 12/13 HKH AIR BL 4"	5,397	12,100	948	529	684	96	189	143	83	32	23
¹⁰² 12/13 HKH GLS STD 2"	54,920	14,780	111,700	37,550	191,300	168,800	424,000	471,100	519,600	258,000	105,900
Air Blank corrected											
¹⁰² 12/13 HKH BLOOD HEAT 1"	367	795	635	111	1,423	229	643	109	134	10	-2
¹⁰² 12/13 HKH BLOOD HEAT 2"	347	1,205	1,120	98	1,438	315	786	103	68	6	-9
¹⁰² 12/13 HKH BLOOD HEAT 3"	357	1,335	916	38	1,498	170	780	98	183	0	-6
¹⁰² 12/13 HKH BLOOD HEAT 4"	437	1,375	1,165	29	1,211	126	736	26	33	0	2
¹⁰² 12/13 HKH BLOOD HEAT 5"	567	1,775	1,275	151	1,347	250	676	43	53	16	-8
¹⁰² 12/13 HKH BLOOD AIR 1"	417	1,005	738	108	1,497	170	698	57	43	14	-3
¹⁰² 12/13 HKH BLOOD AIR 2"	467	1,185	579	83	1,328	247	630	49	81	3	-5
¹⁰² 12/13 HKH BLOOD AIR 3"	377	2,245	704	81	1,299	356	698	50	97	15	-1
¹⁰² 12/13 HKH BLOOD AIR 4"	407	1,885	610	140	1,677	243	808	123	180	6	-3
¹⁰² 12/13 HKH BLOOD AIR 5"	357	2,145	969	- 139	1,586	136	761	59	103	8	6
Normalized to Ba											

APPENDIX EXPERIMENT 18

Isotope - Raw Counts	Yb 174	Hf 178	Hg 202	Ti 205	Pb 208	Th 232	U 238
²⁰² M/2/13 HKH GLS STD 1"	100,400	72,560	172	11,630	55,260	84,200	98,260
²⁰² M/2/13 HKH AIR BL 1"	14	18	108	17	267	10	14
²⁰² M/2/13 HKH AIR BL 2"	14	8	85	14	153	12	7
²⁰² M/2/13 HKH BLOOD HEAT 1"	10	31	799	15	1,415	10	203
²⁰² M/2/13 HKH BLOOD HEAT 2"	18	30	1,026	17	1,200	15	278
²⁰² M/2/13 HKH BLOOD HEAT 3"	18	32	1,139	23	1,840	26	382
²⁰² M/2/13 HKH BLOOD HEAT 4"	9	39	561	12	1,389	15	163
²⁰² M/2/13 HKH BLOOD HEAT 5"	20	53	538	16	1,331	14	219
²⁰² M/2/13 HKH BLOOD AIR 1"	11	30	864	14	1,397	15	125
²⁰² M/2/13 HKH BLOOD AIR 2"	14	53	617	15	1,269	12	211
²⁰² M/2/13 HKH BLOOD AIR 3"	19	50	832	12	1,755	18	134
²⁰² M/2/13 HKH BLOOD AIR 4"	18	67	485	15	1,785	23	407
²⁰² M/2/13 HKH BLOOD AIR 5"	22	68	483	15	1,367	18	188
²⁰² M/2/13 HKH MATRIX BL"	14	97	185	18	1,344	19	378
²⁰² M/2/13 HKH BLOOD 1" no matrix	14	17	1,010	11	1,602	9	9
²⁰² M/2/13 HKH BLOOD 2" no matrix	15	17	1,178	30	1,316	14	10
²⁰² M/2/13 HKH AIR BL 3"	14	15	232	13	157	17	5
²⁰² M/2/13 HKH AIR BL 4"	13	18	209	12	143	11	17
²⁰² M/2/13 HKH GLS STD 2"	108,300	74,610	281	6,293	47,660	87,230	98,340
Air Blank corrected							
²⁰² M/2/13 HKH BLOOD HEAT 1"	-3	15	640	2	1,280	-2	192
²⁰² M/2/13 HKH BLOOD HEAT 2"	4	14	868	4	1,045	4	265
²⁰² M/2/13 HKH BLOOD HEAT 3"	2	15	981	9	1,885	14	302
²⁰² M/2/13 HKH BLOOD HEAT 4"	-4	23	402	-2	1,235	3	153
²⁰² M/2/13 HKH BLOOD HEAT 5"	6	37	380	2	1,238	3	208
²⁰² M/2/13 HKH BLOOD AIR 1"	-2	14	706	1	1,242	3	114
²⁰² M/2/13 HKH BLOOD AIR 2"	1	37	459	2	1,114	1	200
²⁰² M/2/13 HKH BLOOD AIR 3"	6	33	674	-1	1,500	6	123
²⁰² M/2/13 HKH BLOOD AIR 4"	4	51	326	1	1,630	12	396
²⁰² M/2/13 HKH BLOOD AIR 5"	8	51	324	2	1,212	7	187
Normalized to Ba							

APPENDIX EXPERIMENT 18

Isotope - Raw Counts	Li 7	Mg 24	Ca 44	V 51	Cr 52	Mn 55	Fe 56	Co 59	Ni 60	Cu 65	Zn 66
⁷⁰ 212/13 HKH BLOOD HEAT 1"	169	52,290	14,815	179	3,115	6,672	3,350,950	251	1,220	2,748	6,536
⁷⁰ 212/13 HKH BLOOD HEAT 2"	-230	45,206	12,622	192	4,688	5,687	3,113,017	246	1,433	2,863	5,718
⁷⁰ 212/13 HKH BLOOD HEAT 3"	-12	45,380	12,033	202	6,632	7,429	2,922,845	266	821	3,282	5,923
⁷⁰ 212/13 HKH BLOOD HEAT 4"	-463	45,213	9,979	218	6,438	5,688	3,341,987	364	997	2,712	6,232
⁷⁰ 212/13 HKH BLOOD HEAT 5"	-361	54,377	13,959	253	5,329	3,839	3,610,816	432	1,133	2,138	7,503
%Stdev	<det limit	9	15	14	27	22	8	27	21	15	11
⁷⁰ 212/13 HKH BLOOD AIR 1"	-761	58,628	15,758	217	7,028	5,434	3,132,643	332	1,751	3,300	7,343
⁷⁰ 212/13 HKH BLOOD AIR 2"	-907	60,737	11,569	268	3,373	3,296	3,968,120	401	2,154	2,248	7,724
⁷⁰ 212/13 HKH BLOOD AIR 3"	-578	77,052	12,357	203	4,894	2,803	3,135,670	408	2,190	1,820	7,362
⁷⁰ 212/13 HKH BLOOD AIR 4"	-516	53,911	9,598	248	5,944	3,474	3,270,755	242	916	2,640	6,233
⁷⁰ 212/13 HKH BLOOD AIR 5"	-440	59,269	10,030	328	6,150	3,487	3,939,361	353	2,130	2,182	6,423
%Stdev	<det limit	14	21	19	25	27	12	19	30	22	9
⁷⁰ 212/13 HKH BLOOD 1" no matrix	5,276	102,900	39,780	245	13,780	5,998	2,779,000	4,441	58,110	5,066	8,127
⁷⁰ 212/13 HKH BLOOD 2" no matrix	5,511	133,500	52,230	267	14,880	6,401	3,997,000	4,568	58,050	7,003	12,500
(Median air blank)	5,990	40,990	26,715	240	11,435	5,304	103,050	4,920	57,110	2,061	1,353
Blank corrected	<dl	61,910	13,065	5	2,345	694	2,675,950	<dl	1,000	3,005	6,775
	<dl	82,510	25,515	27	3,445	1,097	3,893,950	<dl	940	4,942	11,148
Normalized to Ba	<det limit	61,910	13,065	5	2,345	694	2,675,950	<det limit	1,000	3,005	6,775
	<det limit	69,211	19,089	20	2,577	821	2,913,224	<det limit	703	3,697	8,340
%Stdev	<det limit	8	26	84	7	12	8	<det limit	25	15	15

APPENDIX EXPERIMENT 18

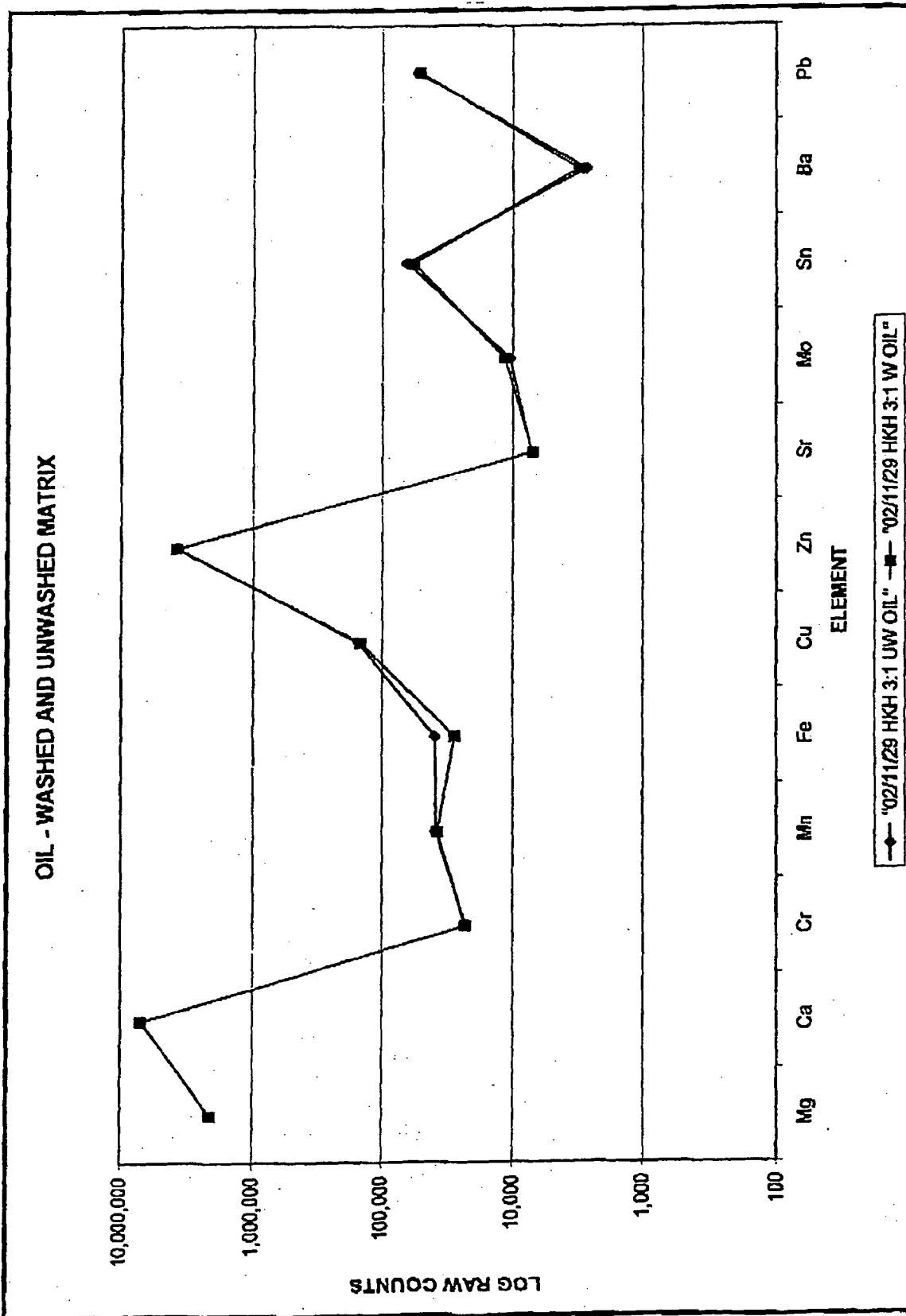
Isotope - Raw Counts	As 75	Se 78	Mo 98	Cd 114	Sn 120	Sb 121	Ba 138	La 139	Ce 140	Eu 151	Dy 162
¹⁰² 12/13 HKH BLOOD HEAT 1"	367	786	635	111	1,423	229	643	109	134	10	-2
¹⁰² 12/13 HKH BLOOD HEAT 2"	284	987	917	80	1,177	258	643	84	56	5	-7
¹⁰² 12/13 HKH BLOOD HEAT 3"	471	1,130	776	32	1,268	144	643	83	155	0	-5
¹⁰² 12/13 HKH BLOOD HEAT 4"	382	1,202	1,019	25	1,058	110	643	23	29	0	2
¹⁰² 12/13 HKH BLOOD HEAT 5"	540	1,691	1,215	144	1,283	238	643	41	51	15	-7
%Stdev	24	29	24	65	11	33	0	52	66	<det limit	<det limit
¹⁰² 12/13 HKH BLOOD AIR 1"	384	926	681	99	1,379	156	643	53	40	13	-3
¹⁰² 12/13 HKH BLOOD AIR 2"	476	1,209	591	85	1,355	252	643	50	83	3	-5
¹⁰² 12/13 HKH BLOOD AIR 3"	348	2,074	651	75	1,200	329	643	47	89	14	-1
¹⁰² 12/13 HKH BLOOD AIR 4"	324	1,501	645	112	1,335	184	643	98	143	5	-2
¹⁰² 12/13 HKH BLOOD AIR 5"	301	1,813	819	118	1,340	115	643	50	87	7	5
%Stdev	19	30	13	18	5	40	0	38	41	<det limit	<det limit
¹⁰² 12/13 HKH BLOOD 1" no matrix	12,770	8,787	899	752	974	270	1,672	190	74	32	18
¹⁰² 12/13 HKH BLOOD 2" no matrix	16,230	11,140	1,138	725	1,268	283	2,175	214	82	34	20
(Median air blank)	4,784	12,585	885	533	705	91	178	119	76	31	19
Blank corrected	<dl	<dl	115	219	270	178	1,494	71	<dl	<dl	<dl
	<dl	<dl	253	192	584	102	1,997	95	<dl	<dl	<dl
Normalized to Ba	<det limit	<det limit	115	219	270	178	1,494	71	<det limit	<det limit	<det limit
	<det limit	<det limit	189	144	422	144	1,494	71	<det limit	<det limit	<det limit
%Stdev	<det limit	<det limit	35	29	31	15	0	1	<det limit	<det limit	<det limit

APPENDIX EXPERIMENT 18

Isotope - Raw Counts	Yb 174	Hf 178	Hg 202	Tl 205	Pb 208	Th 232	U 238
'02/12/13 HKH BLOOD HEAT 1"	-3	15	640	2	1,260	-2	192
'02/12/13 HKH BLOOD HEAT 2"	4	11	710	3	856	3	217
'02/12/13 HKH BLOOD HEAT 3"	2	13	830	8	1,427	12	298
'02/12/13 HKH BLOOD HEAT 4"	-4	20	352	-2	1,079	3	133
'02/12/13 HKH BLOOD HEAT 5"	6	35	362	2	1,178	3	188
%Stdev	<det limit	51	37	<det limit	18	<det limit	29
'02/12/13 HKH BLOOD AIR 1"	-2	13	650	1	1,145	3	105
'02/12/13 HKH BLOOD AIR 2"	1	37	468	2	1,137	1	204
'02/12/13 HKH BLOOD AIR 3"	5	31	622	-1	1,478	6	114
'02/12/13 HKH BLOOD AIR 4"	3	40	260	1	1,288	10	315
'02/12/13 HKH BLOOD AIR 5"	7	43	274	2	1,025	6	158
%Stdev	<det limit	37	41	<det limit	14	<det limit	48
'02/12/13 HKH BLOOD 1" no matrix	14	17	1,010	11	1,602	9	9
'02/12/13 HKH BLOOD 2" no matrix (Median air blank)	15	17	1,178	30	1,316	14	10
	14	16	158	14	155	11	11
Blank corrected	<dl	<dl	852	<dl	1,447	<dl	<dl
	<dl	<dl	1,020	<dl	1,161	<dl	<dl
Normalized to Ba	<det limit	<det limit	852	<det limit	1,447	<det limit	<det limit
	<det limit	<det limit	763	<det limit	869	<det limit	<det limit
%Stdev	<det limit	<det limit	8	<det limit	35	<det limit	<det limit

APPENDIX EXPERIMENT 13

Isotope - Raw Counts	Mg 24	Ca 44	Cr 52	Mn 55	Fe 56	Cu 65	Zn 66	Sr 88	Mo 98	Sn 128	Ba 138	Pb 207
"02/11/29 HKH GLS STD 1"	94,550	631,500	134,200	203,300	210,500	36,830	21,900	378,700	98,200	145,300	302,700	12,200
"02/11/29 HKH GLS STD 2"	105,400	687,700	151,700	233,900	235,200	43,820	25,290	434,100	113,900	175,000	358,300	16,610
"02/11/29 HKH AIR BL 1"	37,290	48,350	2,361	4,460	38,320	2,936	361	555	276	315	87	23
"02/11/29 HKH AIR BL 2"	34,630	41,380	2,390	4,175	34,240	2,682	347	532	272	254	94	23
"02/11/29 HKH 3:1 UW BL"	62,890	49,770	4,236	5,866	159,200	3,022	6,574	1,775	539	1,589	2,326	2,748
"02/11/29 HKH 3:1 W BL"	54,710	48,510	4,833	5,177	168,200	3,339	6,135	1,899	561	1,749	1,684	2,678
"02/11/29 HKH 3:1 UW OIL"	1,717,000	198,000	23,600	45,040	185,800	49,350	1,055,000	7,619	3,083	22,850	4,233	14,150
"02/11/29 HKH 3:1 W OIL"	1,691,000	198,300	24,160	43,490	194,000	48,220	1,081,000	7,676	3,340	20,840	3,879	13,620
Matrix blank corrected												
"02/11/29 HKH 3:1 UW OIL"	1,654,110	149,230	19,364	39,174	36,500	45,328	1,048,428	5,844	2,545	21,261	1,907	11,402
"02/11/29 HKH 3:1 W OIL"	1,636,290	149,790	19,327	38,313	25,800	44,881	1,074,865	5,777	2,779	19,091	2,195	10,942
Element - Raw Counts	Mg	Ca	Cr	Mn	Fe	Cu	Zn	Sr	Mo	Sn	Ba	Pb
"02/11/29 HKH 3:1 UW OIL"	2,093,810	7,006,103	23,107	39,174	39,913	150,416	3,757,799	7,075	10,558	85,218	2,650	54,012
"02/11/29 HKH 3:1 W OIL"	2,071,253	7,032,394	23,063	38,313	28,135	145,718	3,852,563	6,994	11,532	59,551	3,061	51,833
% Std dev.	0.8	0.3	0.1	1.5	24.5	2.2	1.8	0.8	6.2	7.6	9.9	2.9



APPENDIX EXPERIMENT 15

Element - Raw Counts	Li	Mg	Ca	V	Cr	Mn	Fe	Ni	Cu	Zn
"02/12/06 HKH GLS STD 1"	47,490	65,250	314,800	91,720	84,220	129,400	187,500	115,900	27,130	16,370
"02/12/06 HKH GLS STD 2"	41,942	57,354	271,565	78,799	70,067	105,356	164,876	107,511	22,207	11,341
"02/12/06 HKH GLS STD 3"	41,018	65,479	274,201	77,534	74,012	122,292	181,008	115,329	25,437	15,406
"02/12/06 HKH GLS STD 4"	40,624	66,151	266,149	78,201	72,201	116,400	174,192	116,401	23,432	14,478
"02/12/06 HKH GLS STD 5"	38,540	62,445	269,884	75,257	72,523	116,193	178,409	107,941	22,457	14,211
"02/12/06 HKH GLS STD 6"	48,258	68,644	316,450	89,011	86,965	129,212	191,707	118,299	25,852	14,902
"02/12/06 HKH GLS STD 7"	46,680	64,516	299,838	81,820	75,276	117,909	176,308	104,553	21,660	13,946
"02/12/06 HKH GLS STD 8"	47,022	63,160	285,341	78,841	76,177	117,169	175,239	103,415	22,190	13,141
"02/12/06 HKH GLS STD 9"	53,517	66,282	369,379	109,351	100,166	152,187	212,044	115,211	31,787	21,203
"02/12/06 HKH GLS STD 10"	38,574	54,486	230,407	68,320	64,894	100,749	163,475	107,654	21,080	11,485
"02/12/06 HKH GLS STD 11"	47,238	64,609	300,688	91,892	80,741	127,156	189,277	116,602	25,975	17,487
Average Glass Standard	44,627	63,414	290,791	83,704	77,931	121,276	181,276	111,804	24,474	14,908
% Std dev.	10	6	12	13	12	11	7	5	12	18
Calium Normalized										
"02/12/06 HKH GLS STD 1"	47,490	65,250	314,800	91,720	84,220	129,400	187,500	116,900	27,130	16,370
"02/12/06 HKH GLS STD 2"	51,307	70,161	332,202	96,394	85,713	128,881	201,691	131,517	27,165	13,874
"02/12/06 HKH GLS STD 3"	48,516	77,449	324,325	91,708	87,541	144,645	214,096	138,411	30,087	18,221
"02/12/06 HKH GLS STD 4"	49,406	78,823	317,132	93,181	86,040	138,698	207,559	138,698	27,921	17,251
"02/12/06 HKH GLS STD 5"	47,537	77,072	332,887	92,825	89,453	143,318	220,068	133,139	27,700	17,528
"02/12/06 HKH GLS STD 6"	49,803	70,845	326,598	91,865	89,753	133,365	197,854	122,092	26,692	16,380
"02/12/06 HKH GLS STD 7"	56,074	77,500	360,182	98,287	90,427	141,638	211,791	125,594	26,020	16,753
"02/12/06 HKH GLS STD 8"	56,314	75,842	341,730	94,421	91,231	140,324	208,889	123,852	26,575	15,737
"02/12/06 HKH GLS STD 9"	45,341	55,309	312,952	92,647	84,864	128,939	179,652	97,611	26,931	17,864
"02/12/06 HKH GLS STD 10"	51,511	72,734	307,667	91,235	86,647	134,541	218,308	143,761	28,150	15,338
"02/12/06 HKH GLS STD 11"	46,494	63,787	295,949	90,444	79,469	125,152	186,295	114,764	25,588	17,211
Average Glass Standard	49,890	71,320	324,222	93,157	86,851	135,354	203,152	125,949	27,257	16,512
% Std dev.	7	10	5	2	4	5	8	10	4	8
Drift corrected air blanks										
"02/12/06 HKH AIR BL 1"	3,684	20,190	11,549	152	2,468	3,047	35,855	63,302	808	327
"02/12/06 HKH AIR BL 2"	3,594	20,611	12,257	184	2,720	3,306	40,498	65,600	821	371
"02/12/06 HKH AIR BL 3"	4,650	23,283	12,023	120	3,043	4,094	42,535	69,616	703	406
"02/12/06 HKH AIR BL 4"	4,398	23,124	11,818	144	3,162	4,058	44,044	70,354	725	423
"02/12/06 HKH AIR BL 5"	4,143	25,557	12,948	161	3,528	4,674	48,968	76,409	857	509
"02/12/06 HKH AIR BL 6"	4,059	25,874	13,325	172	3,369	4,495	47,950	76,205	875	454
"02/12/06 HKH AIR BL 7"	4,481	22,498	12,679	172	3,113	4,039	42,523	63,628	782	420
"02/12/06 HKH AIR BL 8"	4,665	21,677	12,652	180	3,087	3,817	42,876	61,853	713	387
"02/12/06 HKH AIR BL 9"	3,888	21,353	11,540	145	2,790	3,535	38,598	68,969	814	395
"02/12/06 HKH AIR BL 10"	3,871	21,358	12,933	182	2,837	3,477	42,447	66,395	853	389
Average	4,083	22,651	12,372	162	3,010	3,854	42,730	68,034	799	406
Element - Raw Counts										

Experiment 15/1

APPENDIX EXPERIMENT 15

Element - Raw Counts	Ga	As	Se	Sr	Zr	Mo	Cd	Sn	Ba	La
"02/12/06 HKH GLS STD 1"	97,640	17,950	5,077	233,800	108,100	64,430	10,920	108,900	235,800	263,700
"02/12/06 HKH GLS STD 2"	80,014	14,411	4,775	203,320	92,902	51,228	8,370	83,587	196,445	226,040
"02/12/06 HKH GLS STD 3"	85,443	14,494	5,615	211,943	98,237	58,704	8,090	94,701	217,337	223,229
"02/12/06 HKH GLS STD 4"	81,691	15,295	5,583	207,032	95,489	54,328	7,412	91,701	195,728	214,196
"02/12/06 HKH GLS STD 5"	84,858	14,524	4,985	200,520	84,666	53,912	7,045	88,138	194,117	211,640
"02/12/06 HKH GLS STD 6"	98,635	18,941	5,688	220,573	105,461	64,205	8,313	100,315	228,030	249,680
"02/12/06 HKH GLS STD 7"	82,557	14,865	5,368	201,842	91,566	53,580	7,146	95,117	199,609	224,992
"02/12/06 HKH GLS STD 8"	83,899	15,447	5,247	193,725	87,960	53,525	7,710	90,454	199,979	212,869
"02/12/06 HKH GLS STD 9"	120,865	20,448	5,202	281,520	131,249	79,054	12,516	131,784	273,378	313,117
"02/12/06 HKH GLS STD 10"	70,750	13,024	4,770	187,271	72,202	48,183	8,450	78,170	170,847	180,676
"02/12/06 HKH GLS STD 11"	97,820	18,164	4,906	228,149	103,497	68,426	11,640	112,414	245,398	268,020
Average Glass Standard	89,479	15,882	5,201	213,609	96,121	59,052	8,582	96,793	214,333	235,271
% Std dev.	14	13	6	13	14	16	22	15	13	15
Cerium Normalized										
"02/12/06 HKH GLS STD 1"	97,640	17,950	5,077	233,800	108,100	64,430	10,920	108,900	235,800	263,700
"02/12/06 HKH GLS STD 2"	97,880	17,829	5,841	248,719	113,646	62,667	10,238	102,252	240,309	276,512
"02/12/06 HKH GLS STD 3"	101,062	17,144	6,641	250,685	116,194	69,435	9,569	112,012	257,065	284,034
"02/12/06 HKH GLS STD 4"	97,339	18,224	6,652	246,691	113,781	64,734	8,832	109,266	233,221	255,228
"02/12/06 HKH GLS STD 5"	104,791	17,915	6,149	247,330	116,765	66,497	8,690	108,714	239,433	281,046
"02/12/06 HKH GLS STD 6"	101,797	17,494	5,870	227,645	108,842	66,263	8,580	103,531	236,373	257,685
"02/12/06 HKH GLS STD 7"	99,171	17,881	6,448	242,484	109,994	64,376	8,565	102,247	239,781	270,273
"02/12/06 HKH GLS STD 8"	100,479	18,500	6,294	232,009	105,342	64,102	9,234	108,328	239,498	254,721
"02/12/06 HKH GLS STD 9"	102,402	17,323	4,407	238,515	111,199	66,986	10,804	111,652	231,616	266,285
"02/12/06 HKH GLS STD 10"	94,480	17,392	6,370	223,375	98,419	64,317	8,613	105,724	228,150	241,276
"02/12/06 HKH GLS STD 11"	96,278	17,877	4,828	224,564	101,868	67,348	11,457	110,643	241,531	283,797
Average Glass Standard	99,393	17,756	5,870	237,799	109,105	65,550	9,676	107,368	238,434	261,232
% Std dev.	3	2	12	4	5	3	11	3	3	3
Drift corrected air blanks										
"02/12/06 HKH AIR BL 1"	280	832	3,019	286	108	284	18	165	122	32
"02/12/06 HKH AIR BL 2"	345	971	3,304	275	128	326	28	182	152	44
"02/12/06 HKH AIR BL 3"	306	908	3,129	320	97	362	20	206	147	38
"02/12/06 HKH AIR BL 4"	315	929	3,241	293	103	353	19	185	153	36
"02/12/06 HKH AIR BL 5"	386	1,091	3,859	314	134	382	25	231	158	46
"02/12/06 HKH AIR BL 6"	388	1,057	4,001	309	122	380	23	223	170	41
"02/12/06 HKH AIR BL 7"	368	939	3,299	286	128	354	28	184	149	40
"02/12/06 HKH AIR BL 8"	368	947	3,228	277	132	350	22	193	156	41
"02/12/06 HKH AIR BL 9"	307	918	2,937	286	113	330	23	189	135	39
"02/12/06 HKH AIR BL 10"	359	994	3,432	278	133	333	23	182	141	41
Average	342	959	3,345	281	120	347	23	196	148	40
Element - Raw Counts										

Experiment 15/2

APPENDIX EXPERIMENT 15

Element - Raw Counts	Co	Eu	Dy	Yb	Hf	Hg	Pb	U
"02/12/06 HKH GLS STD 1"	305,900	145,300	57,670	61,330	42,160	367	36,940	54,670
"02/12/06 HKH GLS STD 2"	250,064	127,020	51,079	52,810	36,002	412	27,794	43,100
"02/12/06 HKH GLS STD 3"	258,624	121,397	47,081	47,634	32,567	525	25,563	43,145
"02/12/06 HKH GLS STD 4"	256,723	114,252	45,268	48,559	31,276	483	25,882	43,881
"02/12/06 HKH GLS STD 5"	248,005	111,211	45,510	45,148	30,669	416	22,459	38,761
"02/12/06 HKH GLS STD 6"	296,397	135,559	53,917	56,454	38,642	426	30,187	54,131
"02/12/06 HKH GLS STD 7"	254,651	121,501	47,787	51,349	35,756	251	26,924	42,254
"02/12/06 HKH GLS STD 8"	255,423	116,918	45,224	47,694	33,289	289	27,444	45,918
"02/12/06 HKH GLS STD 9"	361,055	165,458	65,438	68,903	47,354	338	34,320	54,089
"02/12/06 HKH GLS STD 10"	229,089	101,413	38,979	40,738	27,482	325	21,044	41,430
"02/12/06 HKH GLS STD 11"	310,798	147,527	56,844	61,549	42,538	421	32,514	60,233
Average Glass Standard	275,155	127,960	50,418	52,833	36,266	387	28,281	47,419
% Std dev.	13	14	14	16	16	20	16	14
Carlson Normalized								
"02/12/06 HKH GLS STD 1"	305,900	145,300	57,670	61,330	42,160	367	36,940	54,670
"02/12/06 HKH GLS STD 2"	305,900	155,382	62,485	64,602	45,142	504	34,001	52,724
"02/12/06 HKH GLS STD 3"	305,900	143,588	55,687	56,341	38,520	621	30,236	51,031
"02/12/06 HKH GLS STD 4"	305,900	136,137	53,938	56,478	37,258	576	30,852	52,287
"02/12/06 HKH GLS STD 5"	305,900	137,172	56,134	55,689	38,186	513	27,715	47,810
"02/12/06 HKH GLS STD 6"	305,900	139,905	55,846	58,264	39,881	439	31,155	56,866
"02/12/06 HKH GLS STD 7"	305,900	145,953	57,405	61,684	42,952	302	32,342	50,758
"02/12/06 HKH GLS STD 8"	305,900	140,023	54,161	57,119	39,868	347	32,868	54,992
"02/12/06 HKH GLS STD 9"	305,900	140,182	56,440	59,225	40,120	287	29,077	45,826
"02/12/06 HKH GLS STD 10"	305,900	135,428	52,053	54,401	36,699	434	28,103	56,325
"02/12/06 HKH GLS STD 11"	305,900	145,202	55,751	60,579	41,868	415	32,002	59,284
Average Glass Standard	305,900	142,207	56,034	58,610	40,242	437	31,380	52,779
% Std dev.	0	4	5	5	8	24	8	7
Drift corrected air blanks								
"02/12/06 HKH AIR BL 1"	11	21	6	9	9	282	85	8
"02/12/06 HKH AIR BL 2"	18	23	12	11	10	302	72	8
"02/12/06 HKH AIR BL 3"	14	23	8	10	9	319	74	10
"02/12/06 HKH AIR BL 4"	13	23	8	9	7	317	63	7
"02/12/06 HKH AIR BL 5"	22	29	12	12	7	453	69	11
"02/12/06 HKH AIR BL 6"	14	22	11	10	10	432	63	4
"02/12/06 HKH AIR BL 7"	11	20	8	9	9	228	62	8
"02/12/06 HKH AIR BL 8"	15	19	8	6	11	223	61	8
"02/12/06 HKH AIR BL 9"	16	25	10	11	8	312	74	10
"02/12/06 HKH AIR BL 10"	14	21	8	11	11	287	69	7
Average	15	23	9	10	9	317	67	8
Element - Raw Counts								

APPENDIX EXPERIMENT 15

Element - Raw Counts	U	Mg	Ca	V	Cr	Mn	Fe	Ni	Cu	Zn
"02/12/06 HKH SVEN OIL BL 2"	3,821	235,018	41,480	687	10,990	5,483	150,553	73,186	1,189	167,148
"02/12/06 HKH SVEN OIL BL 3"	3,888	201,744	39,846	683	8,118	5,682	157,177	73,459	2,225	143,782
"02/12/06 HKH SVEN OIL WED 1"	3,742	190,075	33,354	584	8,467	9,138	361,619	71,368	4,519	137,849
"02/12/06 HKH SVEN OIL WED 2"	4,128	196,768	34,940	711	10,163	6,968	268,814	74,881	3,343	143,612
"02/12/06 HKH SVEN OIL THUR 1"	4,719	276,925	62,367	745	11,550	11,882	568,657	81,666	7,485	182,213
"02/12/06 HKH SVEN OIL THUR 2"	4,824	238,792	45,529	1,031	13,952	10,300	534,454	81,080	10,451	176,523
"02/12/06 HKH SVEN OIL FRI 1"	4,810	286,334	68,590	2,446	16,629	19,376	529,987	77,330	18,538	221,004
"02/12/06 HKH SVEN OIL FRI 2"	5,029	238,801	45,334	1,105	13,936	10,525	506,588	83,148	16,947	188,439
"02/12/06 HKH JOHN OIL WED 1"	5,385	580,487	55,967	346	13,776	19,958	234,195	82,858	20,828	304,144
"02/12/06 HKH JOHN OIL WED 2"	5,147	604,376	60,976	417	18,936	22,912	306,614	86,485	20,456	314,960
"02/12/06 HKH JOHN OIL THUR 1"	4,518	408,802	44,199	448	13,941	16,549	270,544	83,824	13,895	212,212
"02/12/06 HKH JOHN OIL THUR 2"	4,282	418,970	45,512	425	14,472	16,970	213,334	83,907	14,674	210,577
"02/12/06 HKH JOHN OIL FRI 1"	4,222	467,862	49,288	415	18,658	18,435	214,237	86,038	15,914	242,640
"02/12/06 HKH JOHN OIL FRI 2"	4,394	455,915	49,408	481	17,280	19,570	285,871	84,323	15,748	265,535
"02/12/06 HKH RYAN OIL WED 1"	5,532	408,850	50,572	619	23,880	10,525	470,647	82,108	5,760	358,710
"02/12/06 HKH RYAN OIL WED 2"	5,315	269,141	37,981	906	17,157	11,958	554,841	87,060	5,272	296,034
"02/12/06 HKH RYAN OIL THUR 1"	5,135	585,490	84,218	607	27,065	15,071	566,053	85,204	8,876	493,578
"02/12/06 HKH RYAN OIL THUR 2"	5,015	413,168	48,800	672	17,325	9,512	387,147	84,519	5,325	391,813
"02/12/06 HKH RYAN OIL FRI 1"	4,885	619,761	67,912	580	24,139	10,701	424,569	85,514	8,871	660,379
"02/12/06 HKH RYAN OIL FRI 2"	5,053	601,154	95,593	588	21,817	11,352	475,080	85,087	7,080	673,978
"02/12/06 HKH DAVE OIL WED 1"	6,284	54,719	49,158	583	14,019	18,012	485,381	82,729	4,151	188,777
"02/12/06 HKH DAVE OIL WED 2"	5,625	53,475	49,934	548	11,867	10,956	418,908	81,447	3,872	168,231
"02/12/06 HKH DAVE OIL THUR 1"	5,731	68,496	61,802	815	12,045	11,243	339,597	83,326	4,070	235,505
"02/12/06 HKH DAVE OIL THUR 2"	5,619	55,528	61,737	606	12,589	9,874	266,282	84,838	4,189	195,804
"02/12/06 HKH DAVE OIL FRI 1"	5,678	97,436	172,212	508	21,079	13,060	357,339	85,922	6,315	200,078
"02/12/06 HKH DAVE OIL FRI 2"	5,618	91,916	162,196	421	19,631	10,788	198,769	85,450	4,682	176,145
"02/12/06 HKH SCOTT OIL WED 1"	7,178	359,173	78,240	921	27,782	98,903	11,839,207	119,587	9,650	1,591,134
"02/12/06 HKH SCOTT OIL WED 2"	6,901	218,524	52,416	820	17,884	52,411	10,702,080	104,254	5,678	1,243,243
"02/12/06 HKH SCOTT OIL THUR 1"	6,355	197,533	50,333	900	18,788	72,574	9,736,842	98,617	8,188	943,194
"02/12/06 HKH SCOTT OIL THUR 2"	6,488	241,759	64,444	1,495	23,479	96,567	13,984,018	111,528	8,980	1,683,237
"02/12/06 HKH SCOTT OIL FRI 1"	6,366	168,149	48,849	1,059	18,013	86,219	8,887,866	101,870	5,466	1,090,938
"02/12/06 HKH SCOTT OIL FRI 2"	6,385	220,839	59,311	1,015	22,930	75,366	10,140,408	109,380	7,714	1,704,562
Average Air Blank Corrected										
Sven Reference Oil										
"02/12/06 HKH SVEN OIL BL 2"	-261	212,467	29,117	525	7,980	1,629	107,823	5,161	396	166,742
"02/12/06 HKH SVEN OIL BL 3"	-195	179,194	27,474	521	5,108	1,828	114,448	5,425	1,433	143,376
Sven Engine Oil										
"02/12/06 HKH SVEN OIL WED 1"	-341	167,524	20,981	432	5,458	5,284	318,890	3,334	3,728	137,443

APPENDIX EXPERIMENT 15

Element - Raw Counts	Ga	As	Se	Sr	Zr	Mo	Cd	Sn	Ba	La
"02/12/06 HKH SVEN OIL BL 2"	24,526	1,977	4,304	1,917	9,882	897	127	919	1,035	82
"02/12/06 HKH SVEN OIL BL 3"	29,525	2,031	4,600	1,662	12,522	676	63	1,128	738	93
"02/12/06 HKH SVEN OIL WED 1"	25,965	1,928	3,601	2,130	4,661	820	56	3,242	3,040	1,147
"02/12/06 HKH SVEN OIL WED 2"	30,868	2,157	3,907	1,631	5,203	795	66	2,728	3,399	1,414
"02/12/06 HKH SVEN OIL THUR 1"	32,120	2,818	5,043	4,285	8,881	795	98	1,527	5,424	1,164
"02/12/06 HKH SVEN OIL THUR 2"	35,587	2,537	4,582	4,238	8,728	1,274	153	3,330	5,778	1,583
"02/12/06 HKH SVEN OIL FRI 1"	37,388	2,604	4,194	7,829	10,680	1,986	84	9,445	4,276	1,476
"02/12/06 HKH SVEN OIL FRI 2"	40,696	2,638	4,626	3,411	18,410	1,195	204	3,850	4,497	855
"02/12/06 HKH JOHN OIL WED 1"	9,870	1,773	3,967	2,911	4,180	2,368	65	11,459	1,955	148
"02/12/06 HKH JOHN OIL WED 2"	12,719	1,820	4,405	3,386	5,247	2,998	64	11,801	2,433	210
"02/12/06 HKH JOHN OIL THUR 1"	20,970	1,731	3,924	2,411	8,571	1,631	60	8,203	1,795	269
"02/12/06 HKH JOHN OIL THUR 2"	19,686	1,807	3,771	2,600	6,313	1,807	36	11,414	1,800	430
"02/12/06 HKH JOHN OIL FRI 1"	19,641	1,859	4,148	2,595	4,743	1,883	49	7,343	1,379	85
"02/12/06 HKH JOHN OIL FRI 2"	18,636	2,021	4,138	2,730	3,164	2,004	85	8,186	1,691	538
"02/12/06 HKH RYAN OIL WED 1"	34,832	1,845	4,188	5,115	1,478	1,855	425	2,205	14,046	186
"02/12/06 HKH RYAN OIL WED 2"	43,453	1,825	4,163	4,187	2,098	1,879	87	2,916	11,678	408
"02/12/06 HKH RYAN OIL THUR 1"	30,594	2,186	5,082	4,212	1,571	1,458	135	3,713	9,008	325
"02/12/06 HKH RYAN OIL THUR 2"	38,900	2,043	4,710	3,311	2,045	1,613	156	4,642	3,163	227
"02/12/06 HKH RYAN OIL FRI 1"	26,133	2,608	4,665	5,494	806	2,030	191	2,728	9,848	206
"02/12/06 HKH RYAN OIL FRI 2"	19,987	2,357	4,752	7,552	1,184	2,647	143	2,640	93,280	211
"02/12/06 HKH DAVE OIL WED 1"	39,625	1,871	3,994	2,142	4,657	1,311	66	3,028	2,242	226
"02/12/06 HKH DAVE OIL WED 2"	38,853	1,977	3,815	2,218	4,073	972	58	3,465	2,100	235
"02/12/06 HKH DAVE OIL THUR 1"	64,661	2,107	4,433	3,038	5,477	2,575	139	2,825	2,087	193
"02/12/06 HKH DAVE OIL THUR 2"	43,001	2,254	4,543	2,689	4,590	1,174	76	1,854	851	101
"02/12/06 HKH DAVE OIL FRI 1"	32,320	2,839	4,719	5,484	3,744	1,265	156	1,603	1,583	108
"02/12/06 HKH DAVE OIL FRI 2"	32,793	2,685	4,653	5,137	3,748	1,220	155	1,657	1,610	110
"02/12/06 HKH SCOTT OIL WED 1"	31,712	2,523	4,503	4,233	8,295	3,284	147	4,314	12,088	116
"02/12/06 HKH SCOTT OIL WED 2"	48,230	2,395	4,437	2,724	9,820	5,003	86	4,241	10,009	124
"02/12/06 HKH SCOTT OIL THUR 1"	48,711	2,660	4,320	2,559	8,751	2,085	88	4,173	11,778	365
"02/12/06 HKH SCOTT OIL THUR 2"	48,863	2,990	4,378	3,483	8,709	4,374	233	6,977	16,437	222
"02/12/06 HKH SCOTT OIL FRI 1"	55,686	3,031	4,616	2,686	11,979	2,139	217	4,371	11,676	280
"02/12/06 HKH SCOTT OIL FRI 2"	44,353	3,122	4,509	3,446	10,427	2,297	158	4,528	14,550	889
Average Air Blank Corrected										
Sven Reference Oil										
"02/12/06 HKH SVEN OIL BL 2"	24,184	1,018	959	1,628	9,742	550	105	723	887	42
"02/12/06 HKH SVEN OIL BL 3"	28,183	1,072	1,255	1,370	12,402	330	40	930	589	53
Sven Engine Oil										
"02/12/06 HKH SVEN OIL WED 1"	25,623	970	258	1,839	4,542	473	34	3,046	2,881	1,107

APPENDIX EXPERIMENT 15

Element - Raw Counts	Ca	Eu	Dy	Yb	Hf	Hg	Pb	U
*02/12/06 HKH SVEN OIL BL 2"	120	32	13	19	103	604	440	82
*02/12/06 HKH SVEN OIL BL 3"	65	27	15	16	33	606	438	102
*02/12/06 HKH SVEN OIL WED 1"	314	26	15	14	97	502	41,988	155
*02/12/06 HKH SVEN OIL WED 2"	108	28	12	19	94	498	43,195	113
*02/12/06 HKH SVEN OIL THUR 1"	197	32	22	14	107	749	66,643	19
*02/12/06 HKH SVEN OIL THUR 2"	673	42	24	22	234	685	65,095	136
*02/12/06 HKH SVEN OIL FRI 1"	528	44	23	29	109	488	77,559	171
*02/12/06 HKH SVEN OIL FRI 2"	945	46	21	25	181	508	59,094	165
*02/12/06 HKH JOHN OIL WED 1"	81	28	10	15	32	876	21,561	53
*02/12/06 HKH JOHN OIL WED 2"	191	30	13	16	74	833	21,248	68
*02/12/06 HKH JOHN OIL THUR 1"	85	24	11	17	110	687	11,754	86
*02/12/06 HKH JOHN OIL THUR 2"	139	26	16	19	122	889	13,188	80
*02/12/06 HKH JOHN OIL FRI 1"	72	25	12	14	24	736	12,871	70
*02/12/06 HKH JOHN OIL FRI 2"	112	23	12	10	107	601	15,171	60
*02/12/06 HKH RYAN OIL WED 1"	300	28	12	18	44	730	13,378	156
*02/12/06 HKH RYAN OIL WED 2"	770	31	19	21	60	778	10,142	190
*02/12/06 HKH RYAN OIL THUR 1"	248	29	18	15	148	1,023	15,181	118
*02/12/06 HKH RYAN OIL THUR 2"	502	40	14	23	58	1,018	10,079	155
*02/12/06 HKH RYAN OIL FRI 1"	395	35	18	42	28	721	9,711	115
*02/12/06 HKH RYAN OIL FRI 2"	233	26	18	21	34	742	11,867	142
*02/12/06 HKH DAVE OIL WED 1"	195	27	15	17	83	450	34,765	160
*02/12/06 HKH DAVE OIL WED 2"	126	25	13	28	82	460	41,522	145
*02/12/06 HKH DAVE OIL THUR 1"	574	25	14	27	78	568	37,894	213
*02/12/06 HKH DAVE OIL THUR 2"	96	78	14	19	33	586	35,358	144
*02/12/06 HKH DAVE OIL FRI 1"	85	27	17	22	17	487	40,138	102
*02/12/06 HKH DAVE OIL FRI 2"	59	27	16	21	18	465	43,944	107
*02/12/06 HKH SCOTT OIL WED 1"	261	29	19	28	181	630	7,987	164
*02/12/06 HKH SCOTT OIL WED 2"	130	29	17	18	44	525	8,630	164
*02/12/06 HKH SCOTT OIL THUR 1"	108	28	16	18	107	608	6,244	198
*02/12/06 HKH SCOTT OIL THUR 2"	95	37	26	22	64	744	7,980	173
*02/12/06 HKH SCOTT OIL FRI 1"	108	35	18	24	114	508	5,951	185
*02/12/06 HKH SCOTT OIL FRI 2"	152	33	18	19	114	639	6,900	151
Average Air Blank Corrected								
Sven Reference Oil								
*02/12/06 HKH SVEN OIL BL 2"	105	9	4	9	94	287	372	74
*02/12/06 HKH SVEN OIL BL 3"	50	5	6	6	24	289	371	94
Sven Engine Oil								
*02/12/06 HKH SVEN OIL WED 1"	300	4	6	4	88	186	41,920	147

APPENDIX EXPERIMENT 15

Element - Raw Counts	Li	Hg	Ca	V	Cr	Mn	Fe	Ni	Cu	Zn
"02/12/06 HKH SVEN OIL WED 2"	45	174,218	22,567	549	7,154	3,114	224,084	6,847	2,550	143,208
"02/12/06 HKH SVEN OIL THUR 1"	636	254,375	49,985	583	8,941	8,008	515,927	13,632	6,692	181,807
"02/12/06 HKH SVEN OIL THUR 2"	741	217,242	33,155	869	10,943	6,446	491,725	13,028	9,658	176,117
"02/12/06 HKH SVEN OIL FRI 1"	727	263,784	58,218	2,284	13,620	15,522	487,257	9,296	17,745	220,598
"02/12/06 HKH SVEN OIL FRI 2"	948	216,051	32,982	943	10,926	6,671	463,856	15,114	16,154	198,033
John Engine Oil										
"02/12/06 HKH JOHN OIL WED 1"	1,302	557,937	43,595	184	10,766	16,102	191,465	14,824	20,035	303,738
"02/12/06 HKH JOHN OIL WED 2"	1,065	581,825	48,603	255	13,826	19,058	263,884	18,451	19,663	314,554
"02/12/06 HKH JOHN OIL THUR 1"	435	387,252	31,826	286	10,931	12,695	227,815	15,780	13,102	211,806
"02/12/06 HKH JOHN OIL THUR 2"	189	396,420	33,139	263	11,462	13,116	170,605	15,873	13,881	218,171
"02/12/06 HKH JOHN OIL FRI 1"	139	445,311	36,915	253	15,648	14,581	171,508	18,004	15,121	242,234
"02/12/06 HKH JOHN OIL FRI 2"	311	433,384	37,037	299	14,281	15,715	243,141	16,288	14,955	265,129
Ryan Engine Oil										
"02/12/06 HKH RYAN OIL WED 1"	1,449	387,300	38,200	457	20,870	6,671	427,918	14,074	4,967	359,304
"02/12/06 HKH RYAN OIL WED 2"	1,232	246,531	25,609	743	14,147	8,105	512,112	19,026	4,479	295,628
"02/12/06 HKH RYAN OIL THUR 1"	1,052	582,939	51,846	445	24,055	11,217	522,323	17,170	8,083	493,112
"02/12/06 HKH RYAN OIL THUR 2"	932	390,615	36,528	510	14,315	5,868	344,417	16,485	4,532	391,207
"02/12/06 HKH RYAN OIL FRI 1"	903	597,211	55,539	397	21,129	6,847	381,839	17,480	8,078	659,972
"02/12/06 HKH RYAN OIL FRI 2"	960	578,604	83,221	425	24,607	7,498	432,361	18,053	6,287	673,571
Dave Engine Oil										
"02/12/06 HKH DAVE OIL WED 1"	2,211	32,168	36,786	420	11,009	14,158	442,652	14,694	3,358	166,371
"02/12/06 HKH DAVE OIL WED 2"	1,542	30,924	37,562	385	8,958	7,101	378,178	13,413	3,079	167,825
"02/12/06 HKH DAVE OIL THUR 1"	1,648	45,948	49,530	652	9,035	7,389	296,868	15,291	3,277	235,088
"02/12/06 HKH DAVE OIL THUR 2"	1,536	32,977	49,365	444	9,580	5,820	223,553	16,804	3,396	195,398
"02/12/06 HKH DAVE OIL FRI 1"	1,595	74,885	159,840	345	18,009	9,205	314,809	17,887	5,522	189,672
"02/12/06 HKH DAVE OIL FRI 2"	1,535	68,365	149,823	259	16,622	6,934	155,040	17,416	3,900	175,740
Scott Engine Oil										
"02/12/06 HKH SCOTT OIL WED 1"	3,096	336,623	65,868	759	24,753	95,049	11,796,478	51,553	8,858	1,590,728
"02/12/06 HKH SCOTT OIL WED 2"	2,818	193,974	40,044	658	14,854	48,557	10,669,351	36,220	4,885	1,242,837
"02/12/06 HKH SCOTT OIL THUR 1"	2,272	174,883	37,961	738	15,778	68,720	9,694,113	31,593	5,385	942,788
"02/12/06 HKH SCOTT OIL THUR 2"	2,405	218,208	52,072	1,333	20,469	92,713	13,941,289	43,494	8,187	1,682,931
"02/12/06 HKH SCOTT OIL FRI 1"	2,273	145,599	36,477	897	15,004	62,365	8,945,136	33,836	4,693	1,090,532
"02/12/06 HKH SCOTT OIL FRI 2"	2,303	198,288	46,938	853	19,921	71,511	10,097,676	41,356	6,921	1,704,156
Average Engine Oil - John										
Average Engine Oil - John	575	467,018	38,519	258	12,836	15,211	211,403	16,538	16,126	299,272
Average Engine Oil - Scott										
Average Engine Oil - Scott	2,528	211,446	46,560	873	18,463	73,152	10,855,674	39,674	6,490	1,375,645

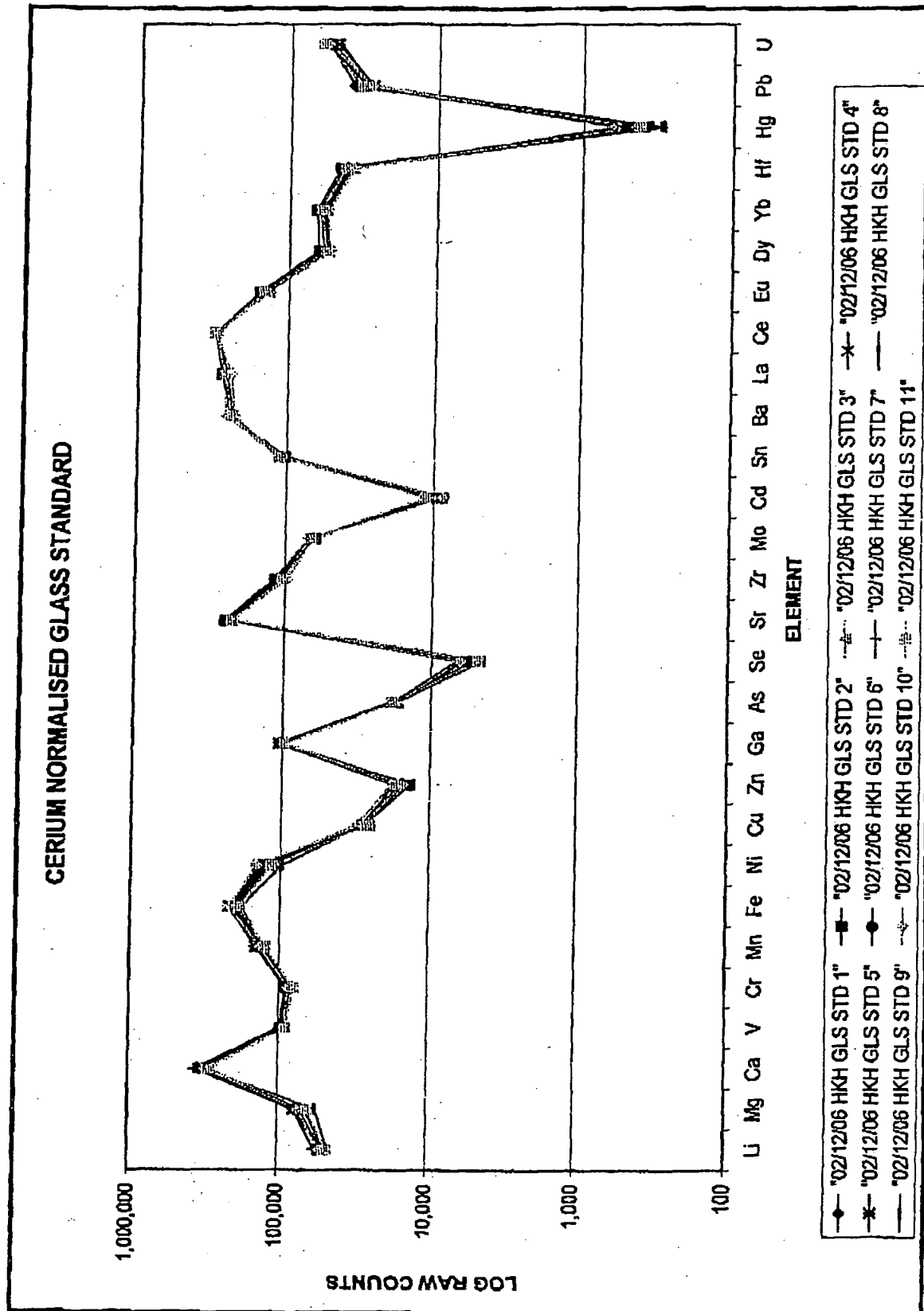
Experiment 15/7

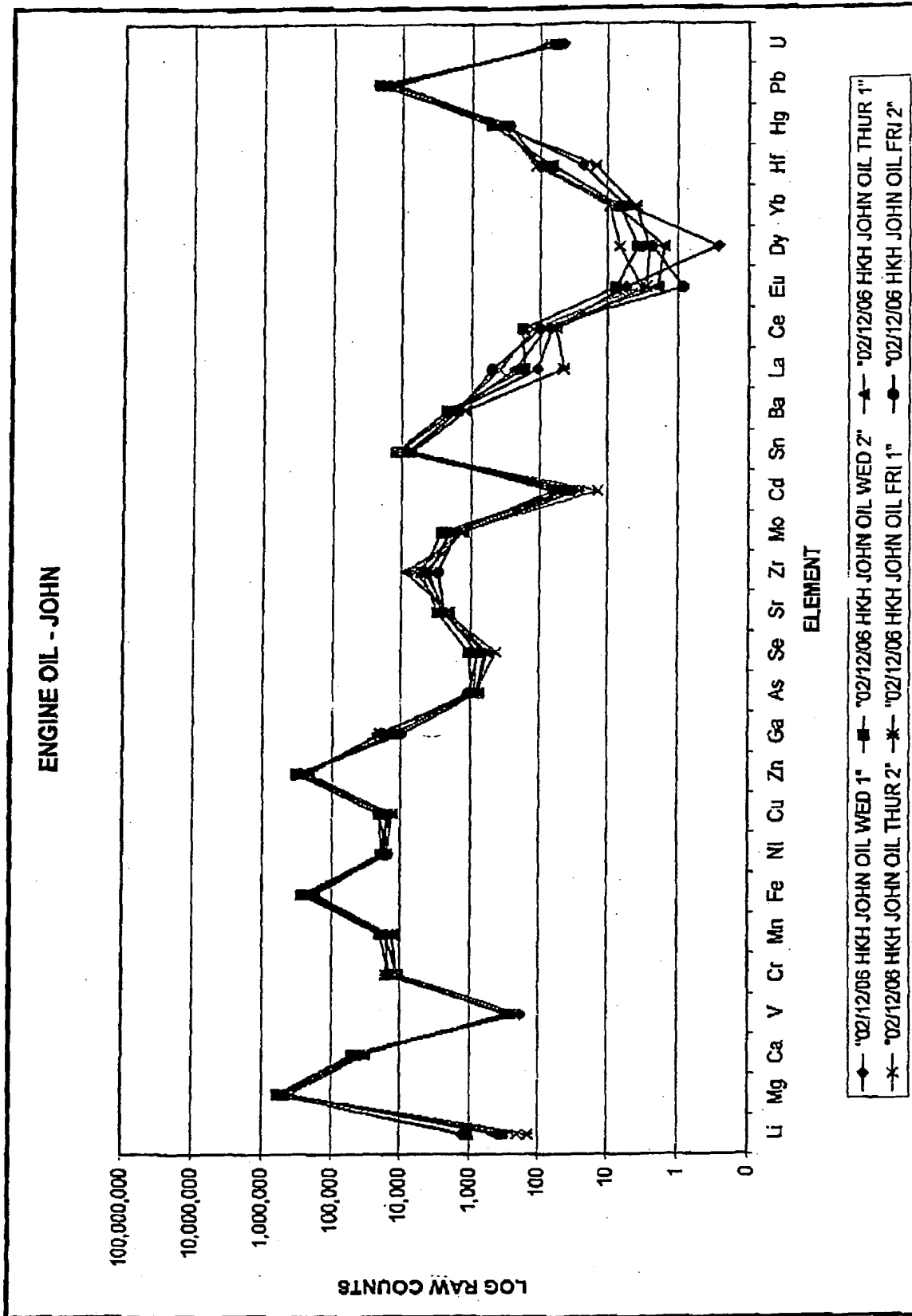
APPENDIX EXPERIMENT 15

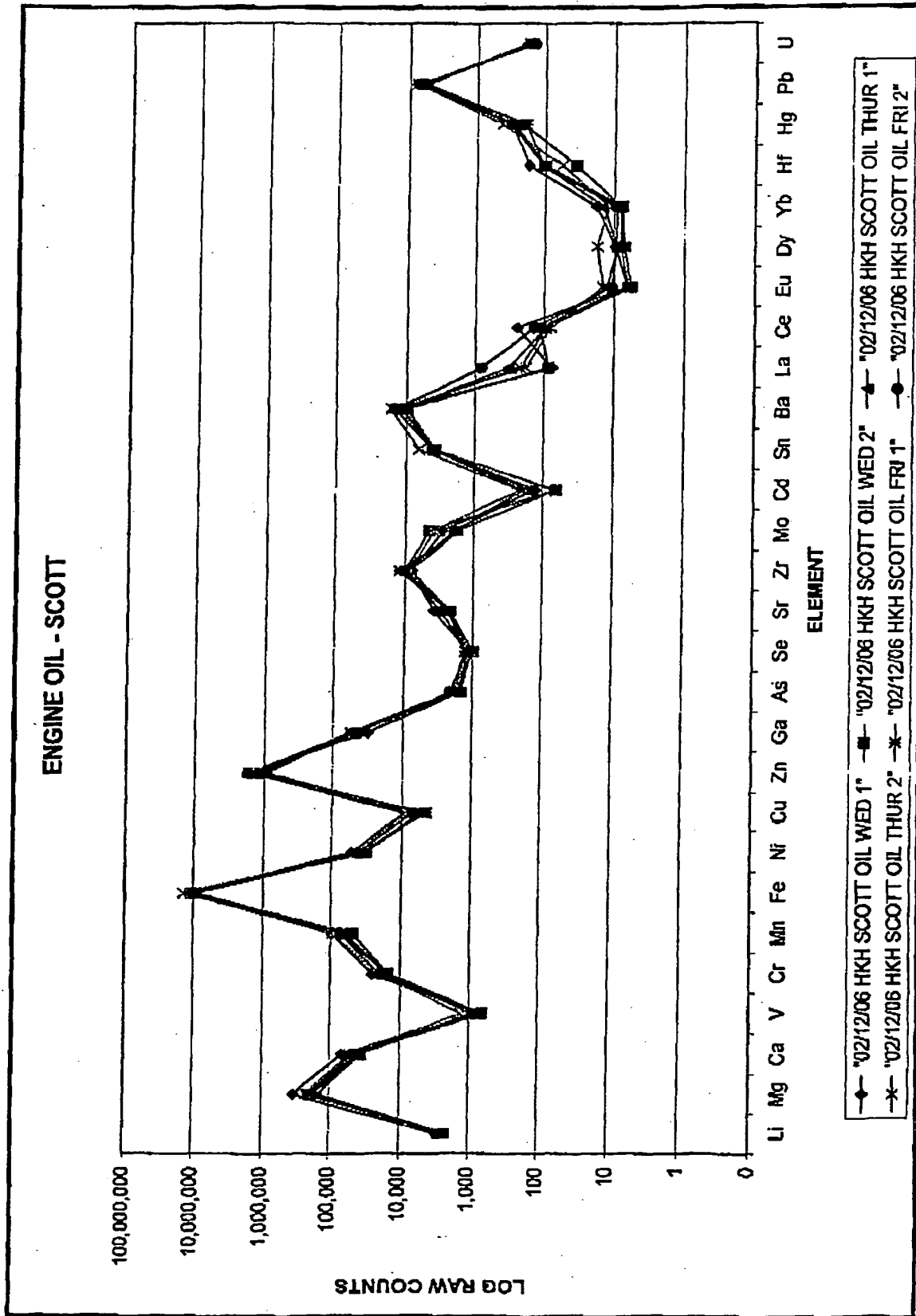
Element - Raw Counts	Ga	As	Se	Sr	Zr	Mo	Cd	Sn	Ba	La
"02/12/06 HKH SVEN OIL WED 2"	30,524	1,198	562	1,340	5,093	448	44	2,533	3,251	1,374
"02/12/06 HKH SVEN OIL THUR 1"	31,778	1,859	1,698	3,994	9,751	1,002	76	1,331	5,275	1,124
"02/12/06 HKH SVEN OIL THUR 2"	36,225	1,579	1,237	3,946	9,109	927	130	3,134	5,629	1,543
"02/12/06 HKH SVEN OIL FRI 1"	37,046	1,646	849	7,638	10,551	1,540	62	9,249	4,127	1,438
"02/12/06 HKH SVEN OIL FRI 2"	40,353	1,580	1,281	3,119	18,231	848	181	3,654	4,348	815
John Engine Oil										
"02/12/06 HKH JOHN OIL WED 1"	9,528	815	622	2,520	4,060	2,022	42	11,263	1,808	108
"02/12/06 HKH JOHN OIL WED 2"	12,377	862	1,051	3,095	5,127	2,651	42	11,606	2,286	170
"02/12/06 HKH JOHN OIL THUR 1"	20,628	773	579	2,119	8,451	1,284	38	8,007	1,647	229
"02/12/06 HKH JOHN OIL THUR 2"	19,344	849	426	2,309	6,194	1,460	14	11,218	1,651	350
"02/12/06 HKH JOHN OIL FRI 1"	19,299	901	803	2,304	4,623	1,336	26	7,147	1,230	45
"02/12/06 HKH JOHN OIL FRI 2"	18,294	1,062	794	2,438	3,044	1,658	63	7,990	1,543	498
Ryan Engine Oil										
"02/12/06 HKH RYAN OIL WED 1"	34,490	886	843	4,823	1,358	1,508	402	2,009	13,898	146
"02/12/06 HKH RYAN OIL WED 2"	43,111	866	818	3,895	1,979	1,533	65	2,720	11,529	369
"02/12/06 HKH RYAN OIL THUR 1"	30,252	1,227	1,747	3,921	1,451	1,111	113	3,517	8,851	288
"02/12/06 HKH RYAN OIL THUR 2"	36,558	1,084	1,365	3,019	1,925	1,267	134	4,446	3,014	187
"02/12/06 HKH RYAN OIL FRI 1"	25,781	1,548	1,311	5,203	708	1,684	168	2,530	9,700	165
"02/12/06 HKH RYAN OIL FRI 2"	19,645	1,398	1,407	7,260	1,066	2,300	121	2,444	93,131	171
Dave Engine Oil										
"02/12/06 HKH DAVE OIL WED 1"	39,283	912	639	1,850	4,538	965	43	2,832	2,093	186
"02/12/06 HKH DAVE OIL WED 2"	38,511	918	470	1,824	3,953	625	35	3,289	1,951	195
"02/12/06 HKH DAVE OIL THUR 1"	64,319	1,148	1,088	2,747	5,357	2,228	117	2,428	1,938	153
"02/12/06 HKH DAVE OIL THUR 2"	42,659	1,285	1,198	2,398	4,470	827	53	1,658	703	61
"02/12/06 HKH DAVE OIL FRI 1"	31,978	1,880	1,374	5,173	3,624	919	134	1,407	1,414	68
"02/12/06 HKH DAVE OIL FRI 2"	32,451	1,907	1,308	4,846	3,528	873	132	1,461	1,461	71
Scott Engine Oil										
"02/12/06 HKH SCOTT OIL WED 1"	31,370	1,565	1,158	3,942	8,175	2,938	125	4,118	11,947	76
"02/12/06 HKH SCOTT OIL WED 2"	47,888	1,435	1,092	2,432	9,700	4,656	64	4,045	9,851	84
"02/12/06 HKH SCOTT OIL THUR 1"	48,369	1,701	978	2,257	8,532	1,719	65	3,977	11,629	325
"02/12/06 HKH SCOTT OIL THUR 2"	48,521	1,942	1,034	3,192	8,589	4,027	210	6,881	16,289	102
"02/12/06 HKH SCOTT OIL FRI 1"	55,344	2,072	1,271	2,395	11,859	1,793	185	4,175	11,527	220
"02/12/06 HKH SCOTT OIL FRI 2"	44,011	2,184	1,164	3,154	10,307	1,951	136	4,333	14,401	819
Average Engine Oil - John	16,578	893	714	2,481	5,250	1,735	37	9,539	1,694	240
Average Engine Oil - Scott	45,917	1,613	1,116	2,897	9,544	2,847	132	4,555	12,608	284

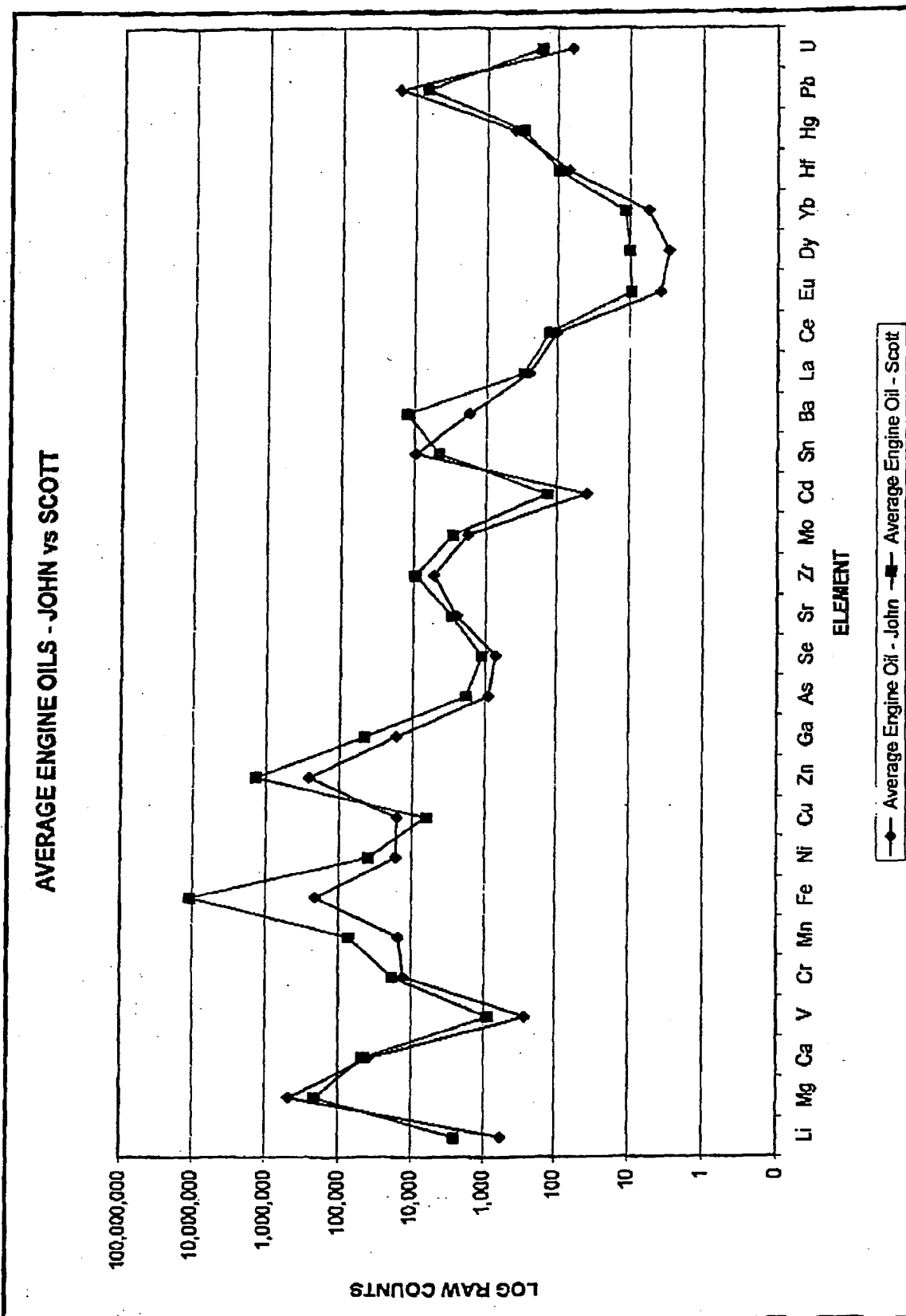
APPENDIX EXPERIMENT 15

Element - Raw Counts	Ce	Eu	Dy	Yb	Hf	Hg	Pb	U
"02/12/06 HKH SVEN OIL WED 2"	93	5	3	9	84	182	43,127	105
"02/12/06 HKH SVEN OIL THUR 1"	182	10	13	4	98	433	68,576	11
"02/12/06 HKH SVEN OIL THUR 2"	658	20	14	12	225	369	65,027	128
"02/12/06 HKH SVEN OIL FRI 1"	511	22	14	19	100	172	77,492	163
"02/12/06 HKH SVEN OIL FRI 2"	930	24	12	15	172	191	59,027	157
John Engine Oil								
"02/12/06 HKH JOHN OIL WED 1"	68	5	0	5	23	359	21,483	45
"02/12/06 HKH JOHN OIL WED 2"	178	7	4	7	65	516	21,181	60
"02/12/06 HKH JOHN OIL THUR 1"	81	2	2	7	109	371	11,886	78
"02/12/06 HKH JOHN OIL THUR 2"	124	3	7	9	112	372	13,121	72
"02/12/06 HKH JOHN OIL FRI 1"	57	3	3	4	15	418	12,803	63
"02/12/06 HKH JOHN OIL FRI 2"	97	1	2	0	98	284	15,103	52
Ryan Engine Oil								
"02/12/06 HKH RYAN OIL WED 1"	285	5	3	9	35	414	13,311	148
"02/12/06 HKH RYAN OIL WED 2"	756	9	10	11	51	463	10,075	182
"02/12/06 HKH RYAN OIL THUR 1"	231	6	7	5	139	706	15,113	111
"02/12/06 HKH RYAN OIL THUR 2"	487	17	5	13	48	701	10,011	147
"02/12/06 HKH RYAN OIL FRI 1"	380	13	7	32	19	405	9,644	107
"02/12/06 HKH RYAN OIL FRI 2"	218	4	8	11	25	426	11,499	134
Dave Engine Oil								
"02/12/06 HKH DAVE OIL WED 1"	180	4	6	7	84	134	34,887	152
"02/12/06 HKH DAVE OIL WED 2"	111	2	4	18	53	143	41,454	137
"02/12/06 HKH DAVE OIL THUR 1"	569	3	5	17	69	252	37,827	205
"02/12/06 HKH DAVE OIL THUR 2"	81	58	5	9	24	279	35,291	136
"02/12/06 HKH DAVE OIL FRI 1"	50	5	7	12	8	170	40,070	94
"02/12/06 HKH DAVE OIL FRI 2"	44	5	7	11	9	149	43,876	99
Scott Engine Oil								
"02/12/06 HKH SCOTT OIL WED 1"	246	6	9	18	172	314	7,919	156
"02/12/06 HKH SCOTT OIL WED 2"	115	8	8	8	35	208	6,563	158
"02/12/06 HKH SCOTT OIL THUR 1"	93	6	7	8	97	292	6,177	190
"02/12/06 HKH SCOTT OIL THUR 2"	80	14	17	12	55	427	7,912	165
"02/12/06 HKH SCOTT OIL FRI 1"	94	12	9	14	105	191	5,894	177
"02/12/06 HKH SCOTT OIL FRI 2"	137	11	9	9	104	322	8,832	143
Average Engine Oil - John	100	4	3	5	69	387	15,898	62
Average Engine Oil - Scott	128	9	10	11	95	282	6,883	164









APPENDIX EXPERIMENT M1

Run	Normalized Data	Blank TE	15/02/2003	7U	986	51V	52Cr	55Mn	59Co	60Ni	66Cu	66Zn	68Ga	75As	82Se	85Rb	88Sr	89Y	90Zr
1				8	0	182	261	42	25	111	23	18	20	18	4	20	21	1	33
2				9	1	184	261	41	24	112	23	18	20	17	4	19	21	1	28
3				8	1	150	263	42	24	110	24	19	20	17	4	18	21	1	20
4				8	0	140	268	42	24	112	24	18	20	18	5	18	22	1	24
5				8	0	132	268	42	24	110	23	19	20	17	4	17	21	1	23
	Mean			8.2	0.5	153.6	263.7	41.8	24.1	111.1	23.4	18.5	18.7	17.0	4.4	18.5	21.3	1.2	26.7
	Standard Deviation			0.2	0.0	19.8	3.0	0.4	0.3	0.8	0.5	0.3	0.2	1.0	0.1	1.3	0.2	0.0	3.7
	Coefficient of Variation			3.0	4.8	12.9	1.1	1.0	1.4	0.7	2.3	1.5	0.8	5.8	1.8	6.8	0.9	4.2	13.9
	Count Limit 3 sigma			N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
6				8	0	128	268	43	23	111	24	19	20	16	4	16	21	1	21
7				8	0	122	264	42	23	110	23	19	20	16	5	16	22	1	20
8				8	1	117	268	42	23	110	24	18	20	16	5	15	21	1	19
9				8	0	111	267	43	23	111	23	18	20	16	5	15	21	1	18
10				7	0	108	269	42	23	110	23	19	20	15	5	15	21	1	18
	Mean			7.8	0.4	118.7	268.5	42.2	23.1	110.8	23.4	18.4	20.1	15.5	4.8	15.6	21.3	1.1	18.1
	Standard Deviation			0.2	0.0	7.5	1.6	0.5	0.3	0.8	0.5	0.5	0.1	0.3	0.1	0.5	0.4	0.1	1.4
	Coefficient of Variation			2.8	10.4	6.4	0.8	1.3	1.4	0.7	2.0	2.4	0.6	2.2	3.2	3.3	1.7	6.7	7.1
	Count Limit 3 sigma			N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
1				24	4	137	377	408	103	113	50	332	80	24	8	105	172	159	96
2				24	5	137	377	405	99	114	49	329	83	24	8	105	174	159	97
3				24	5	135	379	401	101	114	49	322	82	24	6	106	174	157	97
4				24	5	137	377	408	101	115	51	332	82	24	8	102	172	158	98
5				24	5	137	378	401	103	114	51	328	82	24	6	106	171	158	40
	Mean			23.7	4.9	138.4	377.7	403.9	101.4	114.2	49.8	328.7	81.8	23.8	5.8	104.8	172.8	157.8	37.8
	Standard Deviation			0.2	0.1	1.1	1.0	2.6	1.7	1.0	0.8	4.2	1.1	0.2	0.1	1.7	1.3	1.3	1.8
	Coefficient of Variation			0.7	2.7	0.8	0.3	0.7	1.7	0.9	1.8	1.3	1.2	1.0	1.9	1.7	0.7	0.8	4.3
	Count Limit 3 sigma			0.02	0.06	0.02	0.01	0.02	0.05	0.03	0.05	0.04	0.04	0.03	0.08	0.05	0.02	0.02	0.13
6				24	5	138	380	403	101	115	50	332	80	24	8	103	175	158	40
7				24	5	135	377	408	100	115	49	310	82	23	8	101	173	153	41
8				23	4	134	371	403	99	113	73	329	81	23	6	104	174	157	43
9				24	5	134	373	404	101	115	48	326	80	24	8	108	173	156	43
10				23	5	134	373	400	99	114	48	327	81	24	6	102	171	150	44
	Mean			23.7	4.5	134.7	374.8	403.7	100.1	114.3	53.7	328.7	80.9	23.5	5.8	103.1	173.2	158.4	42.2
	Standard Deviation			0.5	0.1	1.2	3.8	2.8	1.0	0.9	10.8	2.3	1.0	0.6	0.2	1.8	1.5	2.5	1.8
	Coefficient of Variation			2.0	1.4	0.8	0.8	0.7	1.0	0.8	20.1	0.7	1.0	2.8	3.4	1.8	0.8	1.6	4.2
	Count Limit 3 sigma			0.06	0.04	0.03	0.03	0.02	0.03	0.02	0.60	0.02	0.03	0.08	0.10	0.05	0.03	0.05	0.13
1				38	8	211	444	585	178	131	69	208	164	34	7	183	282	307	80
2				38	8	211	452	555	173	130	68	203	183	33	7	185	287	312	81

APPENDIX EXPERIMENT M1

Run	Normalized Data	Blank TE	1502/2003	93Mg	93Mg	111Cd	120Sn	121Sb	126Te	138Ba	140Ce	141Pr	146Nd	152Eu	157Gd	158Tb	163Dy	169Ho
1		77	5	0	7	1	1	1	1	1	1	0	0	1	0	1	0	0
2		62	5	0	7	1	1	1	1	1	1	0	0	1	0	0	0	0
3		53	5	0	6	1	1	1	1	1	1	1	0	1	0	1	0	1
4		47	5	0	6	1	1	1	1	1	1	1	0	1	0	1	0	1
5		41	4	0	6	1	1	1	1	1	1	1	0	1	0	1	0	1
	Mean	55.7	4.7	0.2	6.3	1.2	0.9	0.9	0.9	0.5	0.7	0.5	0.2	0.7	0.3	0.5	0.2	0.5
	Standard Deviation	14.0	0.5	0.0	0.8	0.0	0.1	0.1	0.1	0.1	0.0	0.1	0.0	0.1	0.0	0.1	0.0	0.1
	Coefficient of Variation	25.2	10.5	19.1	10.2	1.8	10.8	1.1	13.8	7.2	21.8	19.2	8.2	10.2	10.2	21.8	16.6	16.6
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
16-Feb-03																		
8		37	4	0	5	1	1	1	1	1	1	0	0	1	0	0	0	0
7		33	4	0	5	1	1	1	1	1	1	0	0	1	0	0	0	0
8		31	4	0	5	1	1	1	1	1	1	0	0	1	0	0	0	0
9		28	3	0	5	1	1	1	1	1	1	0	0	1	0	0	0	0
10		28	4	0	5	1	1	1	1	1	1	0	0	1	0	0	0	0
	Mean	31.4	3.6	0.2	5.1	1.0	0.5	0.5	0.5	0.4	0.5	0.3	0.1	0.8	0.2	0.3	0.1	0.4
	Standard Deviation	3.7	0.2	0.0	0.4	0.1	0.0	0.1	0.1	0.1	0.1	0.1	0.0	0.1	0.0	0.1	0.0	0.1
	Coefficient of Variation	11.9	8.8	24.5	7.1	9.8	9.4	0.7	23.7	15.0	18.2	18.2	20.2	12.2	10.3	28.6	27.0	20.9
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
0.1ppm 15/02/2003																		
1		115	41	17	72	62	8	528	188	188	188	238	42	144	45	276	71	274
2		115	39	16	71	61	8	533	188	188	188	236	43	142	45	271	70	279
3		114	38	16	72	61	8	557	183	183	181	233	43	142	45	289	88	277
4		118	40	17	73	61	8	574	187	187	185	229	43	140	46	270	71	280
5		118	40	18	72	61	8	571	188	188	188	238	43	140	45	271	70	274
	Mean	118.1	39.8	18.4	72.1	61.5	8.0	552.4	188.0	188.0	186.5	235.1	42.7	141.4	45.4	271.4	70.0	278.9
	Standard Deviation	1.8	0.7	0.4	0.5	0.6	0.3	21.1	2.2	2.2	3.5	4.0	0.2	1.9	0.2	2.9	1.3	2.6
	Coefficient of Variation	1.5	1.8	2.4	0.7	0.9	3.3	3.8	1.2	1.2	1.9	1.7	0.4	1.3	0.4	1.0	1.9	0.9
	Count Limit 3 sigma	0.05	0.05	0.07	0.02	0.03	0.10	0.11	0.04	0.06	0.06	0.05	0.01	0.04	0.01	0.03	0.06	0.03
16-Feb-03																		
6		118	39	18	72	61	8	576	188	188	184	237	44	144	45	267	70	275
7		112	40	17	73	61	8	584	188	188	183	233	44	140	44	289	70	272
8		114	40	16	72	60	8	573	185	185	188	237	42	143	44	268	87	275
9		114	38	16	72	60	8	571	187	187	184	231	43	141	45	289	88	278
10		114	40	18	73	62	8	568	184	184	184	230	42	142	44	268	68	268
	Mean	114.3	39.4	18.2	72.4	60.8	7.8	574.0	188.0	188.0	184.5	233.7	42.9	141.9	44.8	268.2	88.9	273.5
	Standard Deviation	2.0	0.4	0.4	0.7	1.1	0.1	6.5	1.5	1.5	1.8	3.2	1.0	1.6	0.5	0.7	1.2	3.7
	Coefficient of Variation	1.8	1.0	2.3	0.9	1.8	1.6	1.1	0.8	0.8	1.0	1.4	2.3	1.2	1.1	0.3	1.7	1.3
	Count Limit 3 sigma	0.05	0.03	0.07	0.03	0.05	0.05	0.03	0.02	0.03	0.03	0.04	0.07	0.03	0.03	0.01	0.05	0.04
0.2ppm 15/02/2003																		
1		219	72	32	135	107	15	404	360	360	358	469	84	281	91	540	135	548
2		215	70	31	134	106	18	405	371	371	382	456	83	281	89	525	138	542

APPENDIX EXPERIMENT M1

Run	Normalized Data	160Er	169Tm	172Yb	175Lu	178Hf	181Ta	182W	205Tl	208Pb	208Bi	232Th	238U
1	Blank TE 15/02/2003	0	1	0	1	49	13	40	3	18	10	33	1
2		0	1	0	1	41	11	43	3	19	8	25	1
3		0	1	0	1	36	9	40	2	19	7	21	1
4		0	1	0	1	34	10	40	2	21	8	18	1
5		0	1	0	1	29	8	34	2	19	5	16	1
	Mean	0.2	0.7	0.2	0.8	37.7	10.4	41.5	2.4	18.2	7.3	22.8	0.8
	Standard Deviation	0.0	0.1	0.0	0.1	7.7	1.6	5.4	0.4	0.7	2.0	8.8	0.1
	Coefficient of Variation	11.2	15.6	28.1	12.4	20.4	15.8	13.1	15.7	3.7	27.4	29.4	17.5
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
6	16-Feb-03	0	1	0	1	27	9	32	2	18	5	14	1
7		0	1	0	1	25	7	30	2	19	4	13	1
8		0	1	0	0	23	8	30	1	18	4	12	1
9		0	0	0	0	21	7	28	2	19	4	11	1
10		0	0	0	0	21	7	27	2	18	3	11	0
	Mean	0.1	0.5	0.1	0.4	23.5	7.9	28.0	1.8	19.0	4.0	12.4	0.5
	Standard Deviation	0.0	0.1	0.0	0.1	2.7	1.0	2.3	0.1	0.4	0.5	1.3	0.0
	Coefficient of Variation	33.1	16.9	21.9	29.2	11.4	12.8	8.1	7.9	2.0	13.0	10.8	5.8
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
1	0.1ppm 15/02/2003	94	281	66	288	43	232	33	185	620	198	208	232
2		93	288	65	284	45	232	35	188	625	198	208	231
3		94	288	67	297	47	238	33	188	622	182	214	239
4		94	280	65	280	48	236	32	184	622	188	212	239
5		92	288	68	286	46	233	33	180	621	183	214	235
	Mean	93.4	280.8	65.9	284.9	45.8	234.0	33.8	188.1	622.1	186.7	211.5	235.2
	Standard Deviation	0.9	4.2	0.9	2.9	1.9	2.7	1.2	2.1	1.7	3.1	2.9	3.5
	Coefficient of Variation	0.9	1.4	1.4	1.0	4.1	1.2	3.8	1.1	0.3	1.6	1.4	1.5
	Count Limit 3 sigma	0.03	0.04	0.04	0.03	0.12	0.03	0.11	0.03	0.01	0.05	0.04	0.04
6	18-Feb-03	91	288	66	286	48	231	32	188	631	194	218	236
7		93	282	67	283	50	230	32	185	623	183	213	235
8		92	288	64	289	50	228	32	183	623	187	220	227
9		93	287	64	281	52	231	31	185	627	199	216	232
10		92	282	64	281	51	229	30	182	608	191	215	230
	Mean	92.2	288.9	66.1	282.3	50.4	229.7	31.4	184.2	622.3	184.7	216.5	232.3
	Standard Deviation	1.2	3.5	1.5	2.6	1.4	1.5	0.7	1.7	8.8	3.1	2.8	3.6
	Coefficient of Variation	1.3	1.2	2.3	0.9	2.8	0.7	2.1	0.9	1.4	1.8	1.3	1.6
	Count Limit 3 sigma	0.04	0.04	0.07	0.03	0.09	0.02	0.08	0.03	0.04	0.05	0.04	0.05
1	0.2ppm 15/02/2003	180	571	129	580	100	431	60	370	846	384	422	488
2		187	566	128	581	102	433	64	387	848	383	428	489

APPENDIX EXPERIMENT M1

Run	Normalized Data	7Li	9Be	51V	52Cr	55Mn	58Co	60Ni	65Cu	66Zn	68Ga	75As	82Se	83Rb	86Sr	88Y	90Zr
3		38	8	212	438	561	173	130	68	206	163	33	7	185	281	305	84
4		38	8	209	437	547	177	132	70	206	161	34	7	190	285	310	85
5		39	8	208	420	556	175	130	72	203	162	33	7	194	283	309	85
	Mean	38.2	8.4	209.8	434.1	554.8	175.4	130.4	69.8	204.9	162.4	33.8	6.8	193.8	285.3	308.4	83.0
	Standard Deviation	0.3	0.4	2.2	8.1	6.7	2.2	0.9	1.5	1.3	1.3	0.4	0.3	2.2	3.6	2.6	2.4
	Coefficient of Variation	0.9	4.2	1.1	2.1	1.2	1.2	0.7	2.2	0.7	0.8	1.2	4.1	1.2	1.3	0.8	2.9
	Count Limit 3 sigma	0.03	0.13	0.03	0.08	0.04	0.04	0.02	0.07	0.02	0.02	0.04	0.12	0.03	0.04	0.03	0.09
18-Feb-03																	
6		38	8	209	405	552	171	130	69	209	181	33	7	183	285	306	88
7		39	8	209	415	549	176	130	67	201	182	33	7	192	277	312	81
8		39	8	209	410	560	174	132	68	198	182	33	7	181	279	302	92
9		38	8	208	404	556	170	130	69	207	165	33	7	188	282	305	95
10		37	8	207	408	559	175	132	68	203	163	33	7	188	279	303	95
	Mean	38.3	8.4	208.1	408.3	553.1	173.2	131.1	68.2	203.4	162.8	32.9	6.9	180.8	280.4	306.0	91.9
	Standard Deviation	0.6	0.3	0.9	4.4	4.3	2.6	1.1	0.8	4.0	1.5	0.3	0.1	2.4	3.3	3.8	3.7
	Coefficient of Variation	1.8	3.3	0.4	1.1	0.8	1.5	0.8	1.2	2.0	0.9	1.0	2.1	1.3	1.2	1.3	4.0
	Count Limit 3 sigma	0.05	0.10	0.01	0.03	0.02	0.04	0.02	0.03	0.06	0.03	0.03	0.08	0.04	0.03	0.04	0.12
1ppm 15/02/2003																	
1		154	39	766	873	1038	737	258	204	280	730	104	16	875	1240	1415	468
2		155	38	771	858	1029	729	254	203	281	738	103	16	887	1250	1430	509
3		155	39	757	850	1040	716	253	202	275	717	103	15	863	1228	1413	517
4		151	38	754	848	1039	722	253	201	277	732	104	16	872	1241	1418	561
5		154	39	759	867	1026	730	253	202	278	719	104	16	868	1245	1421	587
	Mean	153.6	38.7	759.5	859.4	1034.4	728.8	254.1	202.8	276.0	726.9	103.7	15.6	868.8	1240.5	1419.5	524.8
	Standard Deviation	1.7	0.6	8.5	10.9	6.3	8.0	2.4	1.2	2.4	8.4	0.7	0.2	8.3	8.9	9.5	34.5
	Coefficient of Variation	1.1	1.5	0.9	1.3	0.6	1.1	0.9	0.6	0.9	1.2	0.8	1.4	1.0	0.7	0.5	6.6
	Count Limit 3 sigma	0.03	0.04	0.03	0.04	0.02	0.03	0.03	0.02	0.03	0.03	0.02	0.04	0.03	0.02	0.01	0.20
16-Feb-03																	
6		150	39	758	858	1007	727	256	206	274	728	104	15	871	1248	1418	560
7		155	39	758	870	1016	725	254	200	282	721	104	16	872	1225	1397	583
8		150	38	763	868	1022	716	256	201	285	704	102	16	858	1231	1414	575
9		158	38	754	850	1025	720	250	203	276	722	104	16	877	1223	1400	577
10		155	38	762	863	1041	727	257	201	272	724	102	16	891	1258	1432	589
	Mean	154.2	38.4	758.4	861.6	1022.3	723.1	254.5	202.0	273.9	719.7	103.3	15.8	871.9	1237.4	1412.2	578.7
	Standard Deviation	2.5	0.6	4.0	8.4	12.8	4.8	2.9	2.4	6.1	8.1	1.2	0.4	8.5	15.1	14.2	14.1
	Coefficient of Variation	1.6	1.6	0.5	1.0	1.5	0.7	1.1	1.2	2.2	1.3	1.2	2.7	1.0	1.2	1.0	2.4
	Count Limit 3 sigma	0.05	0.05	0.02	0.03	0.04	0.02	0.03	0.04	0.07	0.04	0.04	0.08	0.03	0.04	0.03	0.07
5ppm 15/02/2003																	
1		757	182	3559	3163	5035	3400	860	818	823	3865	462	59	4341	6338	7240	3053
2		745	188	3559	3124	5065	3388	861	801	815	3657	464	60	4408	6238	7247	3051
3		744	185	3558	3128	4988	3319	870	821	826	3872	469	58	4327	6239	7147	3087
4		758	183	3604	3134	4915	3464	853	807	817	3872	458	59	4318	6208	7174	3096
5		750	188	3568	3134	4955	3408	855	825	822	3617	463	58	4333	6383	7209	3141

APPENDIX EXPERIMENT M1

Run	Normalized Data	83Rb	86Rb	117Cd	120Sn	121Sb	126Te	138Ba	139La	140Ca	141Pr	148Nd	153Eu	157Gd	159Tb	163Dy	165Ho
3		214	72	32	134	109	18	413	384	358	458	85	279	89	531	135	548
4		212	71	32	135	108	15	408	385	358	450	83	278	89	532	136	541
5		214	68	32	135	108	16	404	387	358	453	83	278	89	523	138	547
	Mean	214.9	70.6	31.7	134.5	107.8	15.3	406.9	385.3	358.2	458.7	83.4	278.1	88.6	530.3	136.6	545.3
	Standard Deviation	2.5	1.3	0.7	0.8	1.0	0.4	3.7	4.0	2.2	7.3	0.8	1.8	0.9	8.6	1.3	3.8
	Coefficient of Variation	1.1	1.8	2.1	0.6	1.0	2.7	0.9	1.1	0.6	1.6	0.9	0.7	1.0	1.2	0.9	0.7
	Count Limit 3 sigma	0.03	0.05	0.08	0.02	0.03	0.08	0.03	0.03	0.02	0.05	0.03	0.02	0.03	0.04	0.03	0.02
18-Feb-03																	
6		212	71	31	133	109	15	409	387	352	455	85	274	88	522	138	542
7		214	68	31	138	108	14	404	358	358	458	85	271	87	518	138	527
8		217	69	31	134	107	15	410	384	358	448	82	278	86	522	136	538
9		212	68	31	134	106	15	421	359	353	457	84	273	86	531	137	537
10		212	70	31	135	107	16	424	365	358	458	83	278	88	516	137	531
	Mean	213.4	69.5	31.2	134.5	107.5	15.1	413.6	382.6	355.9	455.2	83.8	274.0	88.9	521.5	138.4	534.6
	Standard Deviation	2.0	0.7	0.3	1.2	1.0	0.5	8.3	4.1	3.3	3.7	1.5	2.4	1.0	8.1	0.5	6.0
	Coefficient of Variation	0.8	1.0	1.0	0.9	1.0	3.0	2.0	1.1	0.9	0.8	1.7	0.9	1.1	1.2	0.4	1.1
	Count Limit 3 sigma	0.03	0.03	0.03	0.03	0.03	0.08	0.08	0.03	0.03	0.02	0.05	0.03	0.03	0.04	0.01	0.03
19pm 15/02/2003																	
1		945	333	146	595	436	70	1397	1722	1681	2127	392	1320	408	2503	638	2580
2		942	327	147	804	443	68	1404	1721	1690	2147	390	1286	418	2520	642	2605
3		929	332	142	800	433	69	1385	1704	1630	2129	385	1307	413	2484	628	2578
4		967	325	148	607	440	70	1430	1682	1688	2171	388	1301	412	2474	640	2566
5		950	332	144	582	437	70	1350	1688	1629	2113	387	1288	411	2456	649	2573
	Mean	948.9	328.7	145.3	596.5	437.8	69.5	1403.2	1701.1	1657.5	2137.2	390.2	1303.9	412.4	2487.4	639.0	2580.0
	Standard Deviation	12.7	5.3	2.3	8.2	3.4	0.8	18.0	23.4	28.6	22.8	6.1	10.1	3.8	24.9	7.6	15.1
	Coefficient of Variation	1.3	1.6	1.6	1.0	0.8	1.2	1.1	1.4	1.7	1.0	1.5	0.8	1.0	1.0	1.2	0.6
	Count Limit 3 sigma	0.04	0.03	0.05	0.03	0.02	0.04	0.03	0.04	0.05	0.03	0.04	0.02	0.03	0.03	0.04	0.02
18-Feb-03																	
6		935	330	142	598	430	69	1421	1688	1651	2166	390	1305	410	2484	638	2587
7		951	326	142	588	430	69	1400	1681	1647	2124	388	1285	411	2489	637	2582
8		951	334	143	596	430	69	1399	1725	1670	2188	391	1312	411	2484	643	2551
9		956	328	147	600	439	68	1474	1684	1645	2147	397	1328	421	2499	643	2519
10		942	326	147	600	435	71	1417	1701	1656	2190	389	1324	414	2512	644	2557
	Mean	946.8	328.9	144.2	598.6	434.6	69.4	1410.2	1695.4	1658.2	2183.0	391.1	1312.3	413.7	2483.5	641.1	2553.0
	Standard Deviation	8.2	2.4	2.6	1.7	4.1	1.0	14.8	18.5	11.8	27.2	3.3	13.1	4.5	15.6	3.2	23.5
	Coefficient of Variation	0.9	0.7	1.8	0.3	0.9	1.4	1.1	1.1	0.7	1.3	0.8	1.0	1.1	0.6	0.5	0.9
	Count Limit 3 sigma	0.03	0.02	0.06	0.01	0.03	0.04	0.03	0.03	0.02	0.04	0.02	0.03	0.03	0.02	0.01	0.03
5pm 15/02/2003																	
1		4787	1581	712	3011	2175	344	7151	8768	8394	11080	1989	6621	2098	13083	3245	13341
2		4821	1589	721	3008	2185	344	7162	8754	8287	11135	1974	6607	2081	12888	3256	14281
3		4810	1591	710	2984	2223	338	7198	8697	8508	10918	1948	6727	2095	12895	3218	13474
4		4756	1580	700	2940	2143	328	7041	8685	8477	11185	1968	6749	2102	13112	3221	13424
5		4720	1577	710	2864	2192	332	7312	8804	8539	11098	1834	6698	2059	12828	3043	13383

APPENDIX EXPERIMENT M1

Run	Normalized Data	1681F	1681Tm	1721b	1731u	1781W	1811a	1821W	2051T	2081Pb	2081Bi	232Th	238U
3		182	554	129	578	105	427	62	362	833	382	433	450
4		181	585	130	575	106	429	70	370	827	384	431	450
5		181	558	128	568	105	423	82	359	818	383	430	455
	Mean	182.3	560.7	128.9	578.1	103.5	426.7	83.8	365.4	834.3	387.2	428.0	455.9
	Standard Deviation	2.8	12.1	0.7	5.2	2.5	3.7	3.9	5.1	11.5	6.0	4.2	6.8
	Coefficient of Variation	1.5	2.1	0.5	0.9	2.4	0.9	6.2	1.4	1.4	1.5	1.0	1.5
	Count Limit 3 sigma	0.05	0.06	0.02	0.03	0.07	0.03	0.19	0.04	0.04	0.05	0.03	0.04
16-Feb-03													
6		183	568	127	561	106	428	69	354	822	386	424	454
7		179	560	126	570	113	425	61	359	818	387	432	457
8		179	561	128	567	112	424	61	368	824	382	430	458
9		180	584	129	570	113	430	63	368	820	378	428	454
10		177	563	130	572	117	432	62	385	841	393	431	444
	Mean	179.7	562.8	128.3	568.2	112.3	430.0	63.1	382.5	825.0	385.4	429.2	452.9
	Standard Deviation	2.0	2.7	1.5	4.1	3.7	50.5	3.1	8.2	9.3	5.1	3.1	5.3
	Coefficient of Variation	1.1	0.5	1.2	0.7	3.3	11.2	5.0	1.7	1.1	1.3	0.7	1.2
	Count Limit 3 sigma	0.03	0.01	0.04	0.02	0.10	0.34	0.15	0.06	0.03	0.04	0.02	0.03
1ppm 15/02/2003													
1		863	2720	603	2738	611	2283	303	1738	1186	1806	2080	2210
2		853	2722	613	2742	619	2325	305	1744	1178	1830	2082	2207
3		860	2688	615	2725	648	2328	308	1884	1179	1816	2112	2145
4		850	2727	616	2738	658	2315	441	1886	1191	1821	2051	2184
5		868	2704	618	2714	674	2312	404	1718	1183	1794	2088	2189
	Mean	860.3	2712.5	611.2	2735.0	641.1	2312.9	351.8	1718.7	1183.3	1811.3	2092.7	2180.3
	Standard Deviation	6.3	15.8	3.7	16.9	25.9	18.2	66.0	25.0	5.3	17.6	21.8	25.8
	Coefficient of Variation	0.7	0.8	0.6	0.6	4.0	0.8	18.8	1.5	0.4	1.0	1.0	1.2
	Count Limit 3 sigma	0.02	0.02	0.02	0.02	0.12	0.02	0.56	0.04	0.01	0.03	0.03	0.04
18-Feb-03													
6		855	2689	611	2738	667	2294	306	1783	1208	1789	2053	2183
7		850	2681	607	2724	674	2287	305	1728	1174	1839	2076	2184
8		855	2725	607	2710	683	2271	300	1794	1172	1776	2089	2150
9		847	2677	608	2717	686	2300	345	1711	1169	1782	2078	2158
10		862	2684	602	2735	678	2283	304	1720	1178	1838	2074	2159
	Mean	854.8	2683.2	607.0	2724.8	677.7	2287.0	312.3	1731.1	1180.1	1804.8	2073.1	2189.0
	Standard Deviation	6.8	18.6	3.2	12.0	7.0	10.9	18.6	19.7	18.0	31.3	13.1	18.8
	Coefficient of Variation	0.8	0.7	0.5	0.4	1.0	0.5	6.0	1.1	1.4	1.7	0.6	0.9
	Count Limit 3 sigma	0.02	0.02	0.02	0.01	0.03	0.01	0.18	0.03	0.04	0.05	0.02	0.03
5ppm 15/02/2003													
1		4352	14247	3083	14951	3580	11584	1572	8857	5921	9318	10930	11280
2		4338	14147	3050	14833	3608	11557	1504	8856	5896	9294	10826	11251
3		4378	14039	3148	14440	3723	11786	1608	8915	6020	9328	10905	11368
4		4327	14571	2894	14726	3699	11433	1559	8769	5882	9324	10775	11284
5		4378	14782	3125	15061	4051	11386	1573	8774	5886	9209	10669	10857

APPENDIX EXPERIMENT M1

Run	Normalized Data	7Li	9Be	51V	52Cr	56Mn	58Co	60Ni	65Cu	68Zn	69Ga	75As	82Se	83Rb	85Sr	89Y	90Zr
1	Mean	750.4	188.1	3571.8	3138.7	4883.1	3534.7	898.8	914.3	820.9	3666.7	463.0	58.9	4344.9	8277.4	7203.4	3087.8
2	Standard Deviation	5.9	2.5	18.6	15.3	80.3	56.8	0.3	10.1	4.9	22.1	4.7	0.0	36.5	68.4	42.9	37.2
3	Coefficient of Variation	0.8	1.3	0.5	0.5	1.2	1.6	0.7	1.1	0.6	0.6	1.0	1.4	0.8	1.1	0.8	1.2
4	Count Limit 3 sigma	0.02	0.04	0.02	0.01	0.04	0.06	0.02	0.03	0.02	0.02	0.03	0.04	0.03	0.03	0.02	0.04
5	18-Feb-03																
6	Mean	757	189	3567	3098	5028	3591	868	916	828	3633	484	59	4375	8383	7238	3170
7	Standard Deviation	754	191	3584	3181	5030	3587	856	827	828	3638	482	60	4386	8286	7276	3210
8	Coefficient of Variation	749	185	3606	3183	5008	3523	850	824	824	3653	457	59	4275	8272	7208	3152
9	Count Limit 3 sigma	752	191	3563	3167	4971	3481	845	903	811	3566	483	60	4302	8204	7268	3147
10	Mean	748	188	3580	3180	4908	3497	839	909	812	3622	480	59	4316	8169	7116	3077
11	Standard Deviation	751.4	188.4	3579.8	3153.9	4968.9	3535.8	851.2	916.4	820.7	3628.3	481.1	59.2	4330.7	8265.0	7231.1	3181.1
12	Coefficient of Variation	4.3	2.8	20.3	32.1	51.1	50.7	10.3	10.1	8.8	36.9	2.8	0.4	47.8	67.5	74.2	48.5
13	Count Limit 3 sigma	0.8	1.5	0.8	1.0	1.0	1.4	1.2	1.1	1.1	1.0	0.6	0.7	1.1	1.1	1.0	1.5
14	18-Feb-03																
15	Mean	1531	372	7229	6163	10888	7201	1804	1832	1332	7371	913	111	8804	12637	15704	8540
16	Standard Deviation	1524	374	7177	6120	11088	7218	1821	1845	1342	7258	914	109	9001	12844	15875	7263
17	Coefficient of Variation	1502	370	7257	6100	11047	7084	1810	1841	1332	7348	913	112	8931	12893	15740	6418
18	Count Limit 3 sigma	1514	365	7167	5991	10849	7092	1806	1868	1329	7209	898	109	8911	12838	15682	6500
19	Mean	1549	371	7202	5977	11031	7077	1882	1819	1332	7421	903	110	8829	12880	15757	8468
20	Standard Deviation	1524.1	370.4	7208.8	6070.3	11020.9	7130.3	1806.5	1841.3	1333.6	7321.7	908.3	110.2	8853.3	12870.1	15831.8	6637.7
21	Coefficient of Variation	17.9	3.3	37.0	82.0	53.8	73.0	10.5	18.6	5.1	85.7	7.3	1.5	78.8	134.9	133.5	352.2
22	Count Limit 3 sigma	1.2	0.8	0.5	1.4	0.5	1.8	0.7	1.0	0.4	1.2	0.8	1.3	0.9	1.0	0.9	5.3
23	18-Feb-03																
24	Mean	1488	378	7188	6051	11055	7064	1867	1821	1313	7401	891	109	8802	12780	15749	7038
25	Standard Deviation	1525	373	7245	5970	10973	7122	1882	1809	1318	7310	890	110	8748	12870	15698	7083
26	Coefficient of Variation	1493	375	7249	6108	11027	6886	1850	1784	1322	7310	898	109	8735	12888	15551	7102
27	Count Limit 3 sigma	1542	369	7187	6131	10724	7109	1813	1823	1301	7285	892	111	8729	12689	15587	8415
28	Mean	1535	369	7264	6139	10719	7150	1867	1842	1325	7343	888	110	8821	12787	15844	6381
29	Standard Deviation	1518.3	372.6	7222.5	6078.8	10899.4	7084.3	1807.8	1817.7	1315.8	7328.9	892.0	109.8	8788.5	12728.3	15023.7	6801.8
30	Coefficient of Variation	22.5	3.4	42.4	78.2	165.4	85.0	17.1	17.8	9.3	44.6	4.1	0.9	41.8	60.5	76.5	384.8
31	Count Limit 3 sigma	1.5	0.9	0.6	1.2	1.5	0.9	1.1	1.0	0.7	0.6	0.5	0.8	0.5	0.5	0.5	5.4
32	18-Feb-03																
33	Mean	878	141	2800	4180	63088	128	180	881	3033	11700	738	15	142867	5478	88252	104528
34	Standard Deviation	861	140	2800	4113	63217	137	182	886	3051	11792	768	14	140280	5503	85183	103077
35	Coefficient of Variation	873	140	2461	4125	61855	142	185	888	3007	11580	788	15	138428	5379	88325	103207
36	Count Limit 3 sigma	865	140	2413	4088	63189	147	188	877	2988	11452	763	15	140051	5328	82818	102587
37	Mean	857	139	2379	4176	61808	151	187	880	3031	11680	784	15	141545	5334	84342	101882
38	Standard Deviation	858.4	140.1	2530.7	4132.4	62658.9	141.2	183.5	880.3	3024.1	11590.9	788.1	14.8	140714.1	5404.0	84388.2	103047.8
39	Coefficient of Variation	6.1	0.7	172.3	35.6	897.2	8.6	2.9	11.8	21.0	157.5	8.8	0.5	1877.8	82.2	1376.4	981.0
40	Count Limit 3 sigma	0.7	0.5	6.8	0.9	1.1	6.1	1.4	1.3	0.7	1.4	1.3	0.2	1.2	1.5	1.5	1.0
41	18-Feb-03																
42	Mean	1488	378	7188	6051	11055	7064	1867	1821	1313	7401	891	109	8802	12780	15749	7038
43	Standard Deviation	1525	373	7245	5970	10973	7122	1882	1809	1318	7310	890	110	8748	12870	15698	7083
44	Coefficient of Variation	1493	375	7249	6108	11027	6886	1850	1784	1322	7310	898	109	8735	12888	15551	7102
45	Count Limit 3 sigma	1542	369	7187	6131	10724	7109	1813	1823	1301	7285	892	111	8729	12689	15587	8415
46	Mean	1535	369	7264	6139	10719	7150	1867	1842	1325	7343	888	110	8821	12787	15844	6381
47	Standard Deviation	1518.3	372.6	7222.5	6078.8	10899.4	7084.3	1807.8	1817.7	1315.8	7328.9	892.0	109.8	8788.5	12728.3	15023.7	6801.8
48	Coefficient of Variation	22.5	3.4	42.4	78.2	165.4	85.0	17.1	17.8	9.3	44.6	4.1	0.9	41.8	60.5	76.5	384.8
49	Count Limit 3 sigma	1.5	0.9	0.6	1.2	1.5	0.9	1.1	1.0	0.7	0.6	0.5	0.8	0.5	0.5	0.5	5.4
50	18-Feb-03																
51	Mean	878	141	2800	4180	63088	128	180	881	3033	11700	738	15	142867	5478	88252	104528
52	Standard Deviation	861	140	2800	4113	63217	137	182	886	3051	11792	768	14	140280	5503	85183	103077
53	Coefficient of Variation	873	140	2461	4125	61855	142	185	888	3007	11580	788	15	138428	5379	88325	103207
54	Count Limit 3 sigma	865	140	2413	4088	63189	147	188	877	2988	11452	763	15	140051	5328	82818	102587
55	Mean	857	139	2379	4176	61808	151	187	880	3031	11680	784	15	141545	5334	84342	101882
56	Standard Deviation	858.4	140.1	2530.7	4132.4	62658.9	141.2	183.5	880.3	3024.1	11590.9	788.1	14.8	140714.1	5404.0	84388.2	103047.8
57	Coefficient of Variation	6.1	0.7	172.3	35.6	897.2	8.6	2.9	11.8	21.0	157.5	8.8	0.5	1877.8	82.2	1376.4	981.0
58	Count Limit 3 sigma	0.7	0.5	6.8	0.9	1.1	6.1	1.4	1.3	0.7	1.4	1.3	0.2	1.2	1.5	1.5	1.0

APPENDIX EXPERIMENT M1

Run	Normalized Data	93Nb	98Mo	111Cd	120Sn	121Sb	127Te	138Ba	139La	140Ce	141Pr	146Nd	153Eu	157Gd	159Tb	163Dy	165Ho
1	Mean	4780.7	1575.6	710.8	2980.8	2163.7	337.4	1788.3	8757.7	8478.0	11079.4	1860.3	6886.8	2088.7	12981.0	3238.5	13610.8
2	Standard Deviation	42.1	14.8	7.7	32.0	28.0	7.0	99.1	79.2	114.5	98.2	21.6	47.0	18.9	128.0	16.4	384.0
3	Coefficient of Variation	0.9	0.9	1.1	1.1	1.3	2.1	1.4	0.9	1.4	0.9	1.1	0.7	0.8	1.0	0.5	2.9
4	Count Limit 3 sigma	0.03	0.03	0.03	0.03	0.04	0.06	0.04	0.03	0.04	0.03	0.03	0.02	0.02	0.03	0.02	0.08
5	18-Feb-03																
6	Mean	4812	1570	685	3033	2182	338	7247	8786	8432	11040	1878	6853	2082	13154	3274	13431
7	Standard Deviation	4795	1587	714	3085	2196	344	7178	8911	8632	11088	1888	6857	2108	12878	3281	13481
8	Coefficient of Variation	4769	1586	725	2979	2176	335	7108	8731	8628	11081	1888	6848	2059	13008	3241	13512
9	Count Limit 3 sigma	4802	1580	730	3053	2178	335	7104	8801	8463	10886	1953	6788	2125	13108	3279	13389
10	Mean	4754	1582	718	3003	2173	337	7284	8847	8465	11119	1899	6852	2087	13736	3278	13412
11	Standard Deviation	4788.5	1583.8	718.5	3026.6	2180.8	337.8	7184.3	8789.1	8522.1	11088.5	1878.1	6881.8	2088.4	13180.3	3272.6	13440.9
12	Coefficient of Variation	23.8	8.7	13.4	35.7	8.9	3.7	80.8	97.7	94.9	81.4	19.6	83.8	21.2	310.2	18.8	47.7
13	Count Limit 3 sigma	0.5	0.5	1.9	1.2	0.4	1.1	1.1	1.1	1.1	0.5	1.0	1.0	1.0	2.4	0.8	0.4
14	10ppm 15/02/2003	0.02	0.02	0.08	0.04	0.01	0.03	0.03	0.03	0.03	0.01	0.03	0.03	0.03	0.07	0.02	0.01
15	Mean	8570	3173	1395	6112	4431	680	15127	18559	19183	24335	4488	13748	4258	27807	7221	28835
16	Standard Deviation	9708	3218	1445	6180	4444	685	14820	19154	19185	24284	4483	13854	4350	28412	7315	28382
17	Coefficient of Variation	9653	3182	1439	6042	4328	650	14633	19085	19081	25060	4492	13841	4223	28269	7279	28707
18	Count Limit 3 sigma	9771	3183	1435	6040	4379	654	14884	18652	19117	24818	4542	14605	4248	28290	7293	28029
19	Mean	9683	3185	1418	6145	4423	654	14858	19085	19082	24917	4476	13720	4159	28387	7122	28588
20	Standard Deviation	9677.3	3180.2	1425.1	6103.7	4413.0	654.2	14840.6	19188.1	19128.5	24822.4	4481.8	13783.2	4249.1	28233.1	7248.2	28684.3
21	Coefficient of Variation	74.0	17.1	19.5	62.3	28.2	8.3	201.9	230.1	48.3	388.7	28.9	382.7	89.8	245.8	77.4	241.2
22	Count Limit 3 sigma	0.8	0.5	1.4	1.0	0.6	1.0	1.4	1.2	0.3	1.3	8.7	2.8	1.6	0.9	1.1	0.8
23	16-Feb-03	0.02	0.02	0.04	0.03	0.02	0.03	0.04	0.04	0.01	0.04	0.02	0.08	0.06	0.03	0.03	0.03
24	Mean	8571	3140	1389	6075	4455	648	14839	19310	19102	24505	4405	13886	4180	27837	6819	28884
25	Standard Deviation	9518	3158	1408	6138	4385	650	14719	19352	19565	24586	4381	14405	4115	27714	7121	28810
26	Coefficient of Variation	9584	3150	1404	6125	4388	650	14808	19091	19052	24972	4389	14582	4140	27548	7108	28478
27	Count Limit 3 sigma	9690	3168	1395	6109	4384	644	14723	19037	18897	24345	4414	14845	4132	28014	7157	28426
28	Mean	9584	3180	1415	5985	4316	648	14755	18975	19487	24712	4475	14282	4195	28039	7143	28539
29	Standard Deviation	9607.3	3190.2	1404.3	6088.3	4378.6	647.9	14788.0	19153.2	18998.8	24880.4	4408.2	14318.3	4152.4	27889.8	7028.3	28543.6
30	Coefficient of Variation	89.8	15.6	7.8	81.8	50.5	2.3	82.6	188.4	231.4	187.7	42.1	383.9	33.6	287.4	230.1	96.3
31	Count Limit 3 sigma	0.7	0.5	0.6	1.0	1.2	0.4	0.8	0.8	1.2	0.8	1.0	2.7	0.8	1.1	3.3	0.3
32	16-Feb-03	0.02	0.01	0.02	0.03	0.03	0.01	0.02	0.03	0.04	0.02	0.03	0.08	0.02	0.03	0.10	0.01
33	Mean	30012	441	24	1213	185	1	80028	108843	194833	27029	16782	233	3500	3718	8087	5485
34	Standard Deviation	30183	458	24	1431	188	1	79824	108804	190565	28556	16142	231	3483	3718	8130	5485
35	Coefficient of Variation	28899	437	24	1204	188	1	78517	108331	189588	28680	16241	228	3463	3602	8025	5388
36	Count Limit 3 sigma	28565	445	23	1185	185	1	80247	106221	191397	28680	16372	228	3448	3683	8185	5500
37	Mean	28855	442	25	1183	184	1	79483	107173	182200	28403	16186	230	3484	3687	8194	5389
38	Standard Deviation	28822.5	444.2	23.9	1245.1	183.7	0.9	79788.0	107134.4	191888.2	28334.2	16338.8	228.9	3477.8	3681.6	8110.3	5440.9
39	Coefficient of Variation	347.1	7.3	0.5	104.5	1.4	0.1	858.9	1070.3	2008.2	234.4	253.9	2.1	21.8	47.9	53.3	57.8
40	Count Limit 3 sigma	1.2	1.7	2.2	8.4	0.7	10.8	1.1	1.0	1.0	0.9	1.8	0.9	0.6	1.3	0.9	1.1

APPENDIX EXPERIMENT M1

Run	Normalized Data	168Er	168Tm	172Tb	175Lu	178Y	181Th	182W	205Tl	208Pb	208Bi	222Th	228U
1	Mean	4354.9	14377.0	3061.9	14802.1	3730.5	11545.7	1583.0	8834.3	5863.4	8294.1	10821.1	11232.3
2	Standard Deviation	23.5	329.6	59.8	238.1	188.6	149.8	21.8	62.0	51.7	49.5	105.4	159.6
3	Coefficient of Variation	0.5	2.3	1.9	1.6	5.1	1.3	1.4	0.7	0.9	0.5	1.0	1.4
4	Count Limit 3 sigma	0.02	0.07	0.08	0.05	0.15	0.04	0.04	0.02	0.03	0.02	0.03	0.04
5	16-Feb-03												
6	Mean	4356	14151	3085	14748	3673	11621	1800	8806	5900	8551	10799	11474
7	Standard Deviation	4328	14252	3068	15103	4179	11809	1828	8825	6028	9278	10879	11438
8	Coefficient of Variation	4416	14630	3043	14809	3753	11501	1621	8872	5911	9253	10877	11210
9	Count Limit 3 sigma	4365	14754	3303	14658	3749	11553	1600	9055	8040	9338	11043	11318
10	Mean	4357	14859	3129	14938	3756	11603	1816	8857	5802	9297	10870	11260
11	Standard Deviation	4370.5	14549.3	3125.2	14971.3	3822.1	11617.4	1813.2	8903.1	5948.5	8343.6	10913.5	11340.0
12	Coefficient of Variation	36.2	340.3	104.5	173.8	202.8	117.0	12.9	83.8	80.7	120.1	90.3	113.4
13	Count Limit 3 sigma	0.8	2.3	3.3	1.2	5.3	1.0	0.8	1.0	1.4	1.3	0.9	1.0
14	16-Feb-03	0.02	0.07	0.10	0.04	0.16	0.63	0.02	0.03	0.04	0.04	0.03	0.03
15	10ppm 15/02/2003												
16	Mean	8721	30208	6885	34387	8454	24217	3889	18134	13381	18532	22131	23473
17	Standard Deviation	9448	29329	6825	36622	7735	24228	3816	18248	13755	19103	22222	23214
18	Coefficient of Variation	9834	29785	6663	30154	7610	24203	3744	18313	13578	18888	22578	23644
19	Count Limit 3 sigma	8520	29272	6722	30697	7894	24214	3768	18120	13321	18067	22488	23540
20	Mean	9428	29888	6761	30240	8370	24249	3784	18154	13510	18650	22714	23718
21	Standard Deviation	9549.8	29654.8	6777.2	30430.0	7972.8	24221.8	3762.2	18183.3	13588.9	18830.0	22428.0	23537.7
22	Coefficient of Variation	125.2	378.9	88.8	235.6	404.9	17.4	44.2	82.9	136.1	242.1	244.0	202.0
23	Count Limit 3 sigma	1.3	1.2	1.3	0.8	5.1	0.1	1.2	0.5	1.0	1.3	1.1	0.8
24	16-Feb-03	0.04	0.04	0.04	0.02	0.15	0.00	0.04	0.01	0.03	0.04	0.03	0.03
25	Mean	9403	28995	6712	30331	7734	24003	3848	18281	13762	18840	22553	23825
26	Standard Deviation	9704	28991	6579	30368	8389	23770	3835	18282	13405	18774	22185	23380
27	Coefficient of Variation	9430	30078	6734	30151	8389	23802	3705	18093	13506	18591	22046	23308
28	Count Limit 3 sigma	9407	30084	6653	30041	8284	23809	3685	18285	13238	18571	22209	23286
29	Mean	9688	30071	6759	30511	8373	23909	3678	18488	13585	18651	22481	23234
30	Standard Deviation	8522.2	30043.4	6883.5	30284.3	8231.8	23894.5	3688.5	18283.8	13485.0	18887.4	22334.8	23188.0
31	Coefficient of Variation	148.8	48.4	67.7	188.0	281.4	108.1	27.2	132.8	193.9	118.7	208.6	247.0
32	Count Limit 3 sigma	1.8	0.2	1.0	0.8	3.4	0.4	0.7	0.7	1.4	0.8	0.8	1.1
33	16-Feb-03	0.05	0.08	0.03	0.02	0.10	0.01	0.02	0.02	0.04	0.02	0.03	0.03
34	SARW 1 15/02/2003												
35	Mean	6015	2898	4425	2813	5823	7200	570	747	2256	303	58245	21244
36	Standard Deviation	6028	2884	4425	2859	5521	7221	565	748	22048	279	58897	21419
37	Coefficient of Variation	5925	2827	4422	2844	5328	7286	554	757	21512	283	58824	21307
38	Count Limit 3 sigma	5985	2854	4434	2859	5228	7163	543	771	22272	251	59784	21844
39	Mean	5978	2814	4388	2830	5118	7267	582	754	21238	256	59188	21438
40	Standard Deviation	5974.1	2850.7	4420.9	2844.6	5363.7	7227.3	582.6	755.2	21824.7	270.7	58807.4	21450.6
41	Coefficient of Variation	51.3	32.1	13.7	21.4	208.5	48.8	8.0	9.6	431.6	21.8	383.3	234.4
42	Count Limit 3 sigma	0.9	1.1	0.3	0.8	3.9	0.7	1.1	1.3	2.0	8.1	0.6	1.1

APPENDIX EXPERIMENT M1

Run	Normalised Data	7Li	8Be	51V	52Cr	58Ni	60Ni	66Cu	68Ga	75As	82Se	88Sr	89Y	90Zr
1	Count Limit 3 sigma	0.02	0.01	0.20	0.03	0.18	0.04	0.04	0.02	0.04	0.09	0.04	0.05	0.03
2	18-Feb-03													
3		871	139	2353	4131	82352	187	901	3072	11852	778	14	5183	93272
4		872	141	2355	4113	82005	184	880	3010	12153	763	14	5183	93272
5		873	142	2347	4171	83173	184	884	3043	11659	762	15	5444	85807
6		874	140	2338	4138	82500	183	885	3045	11655	778	15	5444	85807
7		875	144	2335	4307	82280	182	880	3043	11823	768	15	5436	84801
8	Mean	871.0	141.2	2342.0	4171.9	82463.9	184.1	890.0	3042.7	11788.5	777.1	14.8	5432	82257
9	Standard Deviation	1.8	1.8	8.0	78.2	435.1	1.7	8.4	21.9	222.8	8.1	0.4	5428.9	10212.6
10	Coefficient of Variation	0.2	1.3	0.3	1.9	0.7	3.1	0.9	0.7	1.9	1.2	2.5	23.3	1189.2
11	Count Limit 3 sigma	0.01	0.04	0.01	0.06	0.02	0.03	0.03	0.02	0.06	0.03	0.07	0.01	0.03
12	SARIM 3 15/02/2003													
13		2716	459	27900	3499	2642853	788	980	21148	23316	331	8	81910	2808788
14		2719	465	27950	3512	2618964	798	981	20859	22818	325	8	83744	2808788
15		2764	484	28032	3552	2598970	813	1003	21653	23207	322	8	82043	2808788
16		2778	470	28088	3520	2620828	815	1005	20888	23428	318	6	82151	2808788
17	Mean	2761	472	27968	3557	2618729	820	1004	21430	23407	315	6	82479	2808788
18	Standard Deviation	2749.6	468.1	27927.5	3527.9	2617845.0	808.5	994.8	21191.8	23314.8	322.4	6.0	82446.4	2808788
19	Coefficient of Variation	25.8	5.1	203.8	25.3	18831.2	10.8	13.0	240.3	364.7	8.3	0.2	784.8	19183.8
20	Count Limit 3 sigma	0.03	0.03	0.02	0.02	0.04	0.02	0.04	0.03	0.05	0.06	0.08	0.02	0.03
21	18-Feb-03													
22		2769	488	28193	3529	2631153	801	998	21288	23286	311	6	82643	2827025
23		2768	472	27960	3543	2634834	823	988	21540	23118	318	6	82747	2845483
24		2787	473	28638	3483	2638253	820	1003	21548	23537	307	6	82821	2782829
25		2827	477	28801	3580	2636882	825	983	21380	23304	308	6	82248	2770510
26	Mean	2758	477	28733	3489	2621253	817	1011	21508	23388	302	6	83087	2827573
27	Standard Deviation	2781.3	473.5	28445.0	3525.2	2637811.1	817.0	998.2	21452.7	23424.4	307.4	6.2	82871.2	2812888.0
28	Coefficient of Variation	27.8	3.8	359.8	40.9	13078.5	8.7	10.9	113.7	250.2	3.4	0.3	304.1	30316.1
29	Count Limit 3 sigma	0.03	0.02	0.04	0.03	0.01	0.04	0.03	0.02	0.04	0.03	0.14	0.03	0.01
30	SARIM 46 15/02/2003													
31		996	17	61357	14481	4089009	21859	8216	44421	325432	5257	13	9143	22031
32		995	17	61476	144517	404171	21881	8036	43410	323332	5080	13	9109	21484
33		977	18	60790	142017	4041842	21690	8992	42585	315842	5002	13	9087	21670
34		981	18	60887	139245	4045884	21747	8883	42943	322298	4888	12	8888	21305
35		1001	18	60818	141077	4077848	21425	8870	43393	323787	5054	12	8891	21305
36	Mean	987.9	16.4	61081.4	142987.5	4087800.8	21840.4	8375.4	43350.8	322533.8	5048.0	12.4	9035.8	21627.3
37	Standard Deviation	9.8	0.7	512.0	2268.2	22955.3	124.8	500.9	889.8	4010.4	140.7	0.2	147.3	289.0
38	Coefficient of Variation	1.0	4.1	0.5	1.6	0.6	0.6	5.3	1.6	1.2	2.8	0.6	1.8	1.2
39	Count Limit 3 sigma	0.03	0.12	0.02	0.05	0.02	0.02	0.16	0.05	0.04	0.08	0.02	0.05	0.04
40	18-Feb-03													

APPENDIX EXPERIMENT M1

Run	Normalized Data	83Nb	88Mo	111Cd	120Sn	121Sb	126Te	138Ba	139La	140Ce	141Pr	146Nd	153Eu	157Gd	163Dy	165Ho
Count Limit 3 sigma		0.03	0.05	0.07	0.25	0.02	0.32	0.03	0.03	0.03	0.03	0.05	0.03	0.02	0.03	0.03
16-Feb-03																
6		28343	441	23	1279	185	1	80420	107747	163925	268970	15989	225	3499	3632	5426
7		28753	442	24	1201	185	1	77820	104333	188026	26217	15907	230	3512	3687	5421
8		30159	447	24	1212	185	1	78162	105605	188740	26053	16176	229	3502	3694	5387
9		28900	438	24	1188	184	1	78633	105523	189171	26202	16258	224	3498	3683	5474
10		30142	441	24	1201	186	1	78604	106357	182158	26882	16198	227	3510	3638	5445
Mean		28658.2	441.9	23.9	1217.9	185.0	0.9	78477.9	105913.4	190400.0	26432.9	16107.7	227.2	3504.2	3688.4	5432.6
Standard Deviation		335.3	3.4	0.6	34.4	0.9	0.0	1052.0	1255.6	2828.5	384.6	148.0	2.5	6.3	30.0	28.8
Coefficient of Variation		1.1	0.8	2.3	2.8	0.5	3.0	1.3	1.2	1.3	1.5	0.9	1.1	0.2	0.8	0.5
Count Limit 3 sigma		0.03	0.02	0.07	0.08	0.01	0.09	0.04	0.04	0.04	0.04	0.03	0.03	0.01	0.02	0.02
SARM 3 : 15/02/2003																
1		358088	207	648	2139	27	10	274441	203870	258146	24057	8321	715	1348	723	827
2		374578	218	634	2159	27	8	274949	204313	257300	25180	10625	709	1356	725	832
3		387379	215	626	2155	28	10	271782	204782	258150	24908	10588	717	1370	726	800
4		306187	215	682	2242	28	8	271715	202131	255880	25280	11020	714	1342	721	827
5		388328	216	658	2210	28	9	271978	207002	256483	25524	10897	718	1374	739	836
Mean		369009.9	213.7	647.2	2180.9	27.6	9.4	272860.8	204418.7	257387.4	25277.3	10488.1	714.2	1357.7	726.8	830.5
Standard Deviation		16463.1	3.6	12.0	43.4	0.7	0.3	1694.3	1757.3	1481.7	284.0	674.8	3.2	14.4	7.3	3.7
Coefficient of Variation		4.1	1.7	1.9	2.0	2.5	3.8	0.8	0.9	0.6	1.1	6.4	0.4	1.1	1.0	0.9
Count Limit 3 sigma		0.12	0.05	0.06	0.06	0.06	0.11	0.02	0.03	0.02	0.03	0.19	0.01	0.03	0.03	0.01
16-Feb-03																
6		376980	216	638	2159	28	9	272727	205041	254853	24708	10789	728	1385	735	831
7		370988	211	653	2155	28	8	275693	205389	255087	25113	10916	718	1378	732	818
8		386330	211	642	2166	27	9	269719	204552	255844	25050	10638	723	1385	738	823
9		384344	208	633	2138	28	9	274209	204408	253612	25104	10822	714	1377	744	831
10		362847	214	635	2157	27	8	271604	202943	251431	25316	10879	718	1361	740	827
Mean		387836.9	212.3	640.1	2152.5	27.4	8.8	273699.4	203906.8	256121.2	25078.3	10888.8	720.4	1378.8	741.8	832.5
Standard Deviation		5428.4	2.9	8.1	8.4	0.4	0.2	3851.3	1074.6	3075.4	165.3	80.1	6.0	12.7	6.3	8.2
Coefficient of Variation		1.5	1.4	1.3	0.4	1.8	1.7	1.1	0.5	1.2	0.7	0.7	0.8	0.9	0.8	0.7
Count Limit 3 sigma		0.04	0.04	0.04	0.01	0.05	0.05	0.03	0.02	0.04	0.02	0.02	0.03	0.03	0.03	0.02
SARM 48 : 15/02/2003																
1		3517	117	3417	1841	250189	3	117294	15682	54588	4226	2881	458	676	584	781
2		3318	114	3421	1780	247832	3	115338	15838	54903	4188	2830	449	688	572	782
3		3245	113	3431	1805	250086	3	115451	15847	54782	4142	2815	435	680	579	778
4		3193	113	3365	1777	247112	2	118788	13775	54613	4149	2856	438	660	575	783
5		3005	114	3478	2342	248927	3	114901	13853	55134	4088	2885	450	673	581	778
Mean		3255.5	114.1	3422.3	1808.2	248911.1	2.6	116148.4	14158.9	54788.9	4153.8	2913.2	448.1	675.0	577.9	777.1
Standard Deviation		186.7	1.7	38.2	243.4	1379.5	0.1	977.9	855.1	223.8	53.8	47.3	9.2	5.8	4.8	5.3
Coefficient of Variation		5.7	1.4	1.1	12.7	0.6	4.9	0.8	6.0	0.4	1.3	1.5	2.1	0.8	0.8	0.7
Count Limit 3 sigma		0.17	0.04	0.03	0.38	0.02	0.15	0.03	0.18	0.01	0.04	0.06	0.06	0.02	0.02	0.02
16-Feb-03																

APPENDIX EXPERIMENT M1

Run	Normalized Data	7L	88e	51V	52Cr	58Mn	58Co	60Ni	65Cu	68Zn	69Ga	75As	82Se	85Rb	86Sr	89Y	90Zr
6		982	16	80435	130650	3904885	21239	8820	42948	319570	4953	35160	13	8702	21401	9531	20164
7		1001	16	59839	136883	4015559	21423	10048	42441	321762	4886	34591	12	8630	21521	9723	19811
8		991	16	60078	140586	4008070	21393	8825	42322	317054	4918	34337	12	8778	21245	9660	19807
9		1000	17	60413	142639	4039688	21471	8919	42927	317286	4901	34499	12	8888	21201	9558	19809
10		1008	16	60264	139638	4026439	21272	8730	42548	317279	4807	34634	12	8781	21577	9733	19802
	Mean	998.5	16.3	60365.3	140078.5	4023820.4	21359.5	9028.8	42637.4	317574.2	4885.1	34844.1	12.0	8825.8	21388.9	9639.1	19888.7
	Standard Deviation	6.8	0.2	372.7	1607.7	25956.9	98.6	580.4	285.6	3556.4	54.3	398.6	0.4	81.5	186.2	92.4	187.4
	Coefficient of Variation	0.7	1.3	0.6	1.1	0.6	0.5	6.4	0.7	1.1	1.1	0.9	3.0	0.9	0.8	1.0	0.9
	Count Limit 3 sigma	0.02	0.04	0.02	0.03	0.02	0.01	0.19	0.02	0.03	0.03	0.03	0.08	0.03	0.02	0.03	0.03
System check 15/02/2003																	
1		794	20.0	3787	3096	4828	3513	889	932	871	3558	492	59	4341	8113	6865	2968
2		808	200	3688	2883	4832	3455	863	925	890	3584	488	58	4331	8181	6866	2960
3		824	201	3683	3190	4914	3501	888	928	851	3573	484	57	4412	8148	7009	3000
4		808	202	3682	3067	4887	3481	866	937	858	3526	479	59	4347	8114	6882	3012
5		802	199	3624	3060	4866	3392	844	916	842	3524	478	56	4283	8028	6879	3015
	Mean	807.0	200.4	3689.1	3071.1	4880.9	3470.1	858.3	927.5	866.4	3548.8	484.2	57.5	4344.8	8130.8	6880.2	2995.0
	Standard Deviation	11.2	1.1	50.8	54.7	59.4	49.0	8.9	7.9	10.7	22.6	5.9	1.0	43.0	53.7	18.5	20.6
	Coefficient of Variation	1.4	0.6	1.4	1.8	1.2	1.4	1.2	0.8	1.2	0.6	1.2	1.7	1.0	0.8	0.3	0.7
	Count Limit 3 sigma	0.04	0.02	0.04	0.05	0.04	0.04	0.03	0.03	0.04	0.02	0.04	0.05	0.03	0.02	0.01	0.02
18-Feb-03																	
6		802	195	3621	3081	4822	3449	850	912	834	3543	472	57	4229	8087	6850	3004
7		788	197	3578	2984	4839	3410	846	908	843	3430	469	57	4206	8083	6802	2988
8		810	197	3583	3003	4783	3444	842	918	840	3469	466	58	4227	8087	6884	2987
9		788	197	3564	2873	4794	3386	850	901	850	3535	488	56	4284	8021	6802	2972
10		777	183	3544	2889	4756	3384	839	907	828	3448	456	55	4193	8025	6814	2977
	Mean	783.0	195.7	3575.8	3010.0	4820.7	3417.1	845.4	908.7	834.9	3486.0	468.1	56.1	4228.5	8018.7	6830.3	2983.8
	Standard Deviation	12.7	1.9	30.1	48.4	64.0	28.4	4.9	6.7	6.4	51.1	6.2	1.0	34.8	50.3	41.7	18.6
	Coefficient of Variation	1.6	1.0	0.8	1.5	1.3	0.8	0.6	0.6	0.8	1.6	1.3	1.8	0.8	0.8	0.8	0.6
	Count Limit 3 sigma	0.05	0.03	0.03	0.03	0.04	0.02	0.02	0.02	0.02	0.04	0.04	0.05	0.02	0.03	0.02	0.02
Blank TE 15/02/2003																	
1		6	1	98	287	45	23	108	25	28	21	18	4	14	25	1	18
2		6	0	96	270	45	23	111	25	30	21	15	5	14	25	1	16
3		7	0	94	289	44	23	114	25	30	21	14	4	13	25	1	14
4		8	0	92	271	44	23	111	26	30	22	15	5	14	26	1	14
5		7	0	90	270	45	23	112	26	30	21	15	5	14	26	1	14
	Mean	7.5	0.4	94.1	289.4	44.4	22.7	111.6	25.4	29.4	21.3	14.9	4.5	13.8	25.6	1.0	15.0
	Standard Deviation	0.2	0.1	3.3	1.9	0.8	0.2	1.7	0.8	0.8	0.3	0.5	0.2	0.3	0.3	0.1	1.0
	Coefficient of Variation	2.4	13.1	3.5	0.6	1.9	1.0	1.5	2.4	2.8	1.4	3.3	4.4	2.2	1.1	8.0	6.6
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
16-Feb-03																	
6		7	1	88	273	43	22	111	27	29	21	14	5	13	25	1	13
7		8	1	86	272	44	23	112	26	29	21	15	6	13	26	1	13
8		7	0	83	283	44	23	113	25	28	20	14	5	12	24	1	13

APPENDIX EXPERIMENT M1

Run	Normalized Data	80Nb	86Mo	111Cd	120Sn	121Sb	125Te	138Ba	139La	140Ce	141Pr	148Nd	153Eu	157Gd	159Tb	163Dy	165Ho
6		3222	111	3547	2378	241899	3	114377	13745	55238	4147	2888	433	677	568	771	571
7		3252	113	3377	243262	3	112985	13528	56163	4051	4088	2859	442	673	570	770	568
8		3131	112	3383	1739	243533	2	115492	13528	56163	4051	2818	436	669	571	767	579
9		3070	110	3365	1753	243570	2	114849	13833	54393	4087	2845	434	663	578	780	569
10		3003	110	3334	2344	246422	3	114205	13460	55172	4088	2859	442	673	570	770	567
	Mean	3135.4	111.1	3391.3	1861.4	244897.3	2.5	113907.6	13560.4	54981.8	4087.0	2851.1	436.8	688.2	568.3	770.7	571.8
	Standard Deviation	106.7	1.3	20.6	337.7	2831.7	0.2	1076.0	188.7	836.2	36.5	24.5	3.5	7.0	4.8	5.4	4.8
	Coefficient of Variation	3.3	1.2	0.6	17.0	1.1	6.2	0.9	1.4	1.5	0.9	0.9	0.8	1.0	0.6	0.7	0.8
	Count Limit 3 sigma	0.10	0.04	0.02	0.51	0.03	0.18	0.03	0.04	0.05	0.03	0.03	0.02	0.03	0.03	0.02	0.02
Sigm check 15/02/2003																	
1		4658	1523	680	2659	2125	318	8588	8489	8109	10389	1905	6340	1988	12422	3142	12778
2		5316	1528	682	2657	2117	322	8590	8398	8241	10552	1889	6409	2008	12430	3155	12874
3		4658	1527	681	2907	2107	327	8600	8464	8284	10595	1852	6344	1969	12384	3140	13010
4		4780	1501	689	2889	2108	322	8619	8457	8111	10382	1921	6408	2005	12387	3156	13072
5		4658	1515	688	2834	2078	324	8465	8278	8118	10590	1888	6400	2040	12742	3156	13207
	Mean	4313.0	1518.9	685.4	2875.3	2107.3	322.5	8532.4	8416.9	8172.4	10540.2	1852.8	6378.8	2001.8	12528.2	3148.0	12875.5
	Standard Deviation	284.5	11.2	5.4	21.9	10.1	3.3	73.6	85.7	83.9	97.4	25.7	34.7	28.3	177.3	8.1	182.8
	Coefficient of Variation	5.9	0.7	0.8	0.8	0.6	1.0	0.8	1.0	1.0	0.9	1.4	0.5	1.3	1.4	0.3	1.3
	Count Limit 3 sigma	0.18	0.02	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.04	0.02	0.04	0.04	0.01	0.04
16-Feb-03																	
6		5375	1525	675	2831	2091	319	8498	8215	8137	10478	1854	6354	1977	12500	3155	13122
7		5334	1486	681	2637	2097	324	8418	8194	8203	10539	1870	6294	1967	12353	3119	12500
8		4582	1503	688	2834	2059	322	8403	8284	8091	10263	1838	6318	1989	12340	3068	12745
9		4580	1485	678	2797	2054	321	8400	8344	8032	10284	1838	6366	1972	12281	3114	12843
10		4704	1481	672	2855	2065	313	8349	8290	8032	10450	1845	6351	1988	12405	3076	12707
	Mean	4935.1	1498.4	674.7	2830.9	2075.3	318.7	8410.8	8253.3	8088.9	10408.9	1852.0	6342.5	1978.2	12371.7	3102.8	12781.6
	Standard Deviation	365.9	17.1	6.5	20.8	18.3	4.4	48.2	63.6	73.2	128.0	11.6	38.4	33	88.3	28.0	222.4
	Coefficient of Variation	7.8	1.1	1.0	0.7	0.9	1.4	0.6	0.8	0.9	1.2	0.6	0.6	0.5	0.7	0.9	1.7
	Count Limit 3 sigma	0.23	0.03	0.03	0.02	0.03	0.04	0.02	0.02	0.03	0.04	0.02	0.02	0.01	0.02	0.03	0.06
Blank TE 15/02/2003																	
1		21	3	0	5	1	0	855	0	0	0	0	1	0	0	0	0
2		18	3	0	5	1	1	850	0	0	0	0	1	0	0	0	0
3		18	3	0	5	1	0	852	0	0	0	0	0	0	0	0	0
4		18	3	0	5	1	0	856	0	0	0	0	0	0	0	0	0
5		18	3	0	5	1	0	801	0	0	0	0	0	0	0	0	0
	Mean	18.6	3.1	0.2	5.3	0.6	0.4	870.7	0.3	0.4	0.1	0.1	0.5	0.2	0.1	0.0	0.1
	Standard Deviation	1.8	0.1	0.0	0.2	0.1	0.1	25.5	0.1	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0
	Coefficient of Variation	9.8	2.9	18.7	4.5	10.3	12.7	2.9	20.3	9.8	23.3	27.2	10.9	24.6	19.3	26.1	31.3
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
16-Feb-03																	
6		16	3	0	6	1	0	877	0	0	0	0	0	0	0	0	0
7		15	3	0	6	1	0	875	0	0	0	0	0	0	0	0	0
8		15	3	0	6	1	0	880	0	0	0	0	1	0	0	0	0

APPENDIX EXPERIMENT M1

Run	Normalized Data	166EJ	169Tm	172Yb	173Lu	178Hf	181Ta	182W	205Tl	208Pb	209Bi	232Th	238U
6		534	215	307	199	589	403	581	221	8147681	9229	9541	1324
7		534	212	314	203	587	405	581	216	8083487	9219	9498	1322
8		532	214	308	200	578	399	578	219	8026488	9219	9288	1303
9		532	220	308	202	588	391	582	219	8030315	9160	9464	1316
10		528	212	308	200	1002	392	1034	219	8104505	9182	9665	1319
	Mean	532.0	214.8	308.7	200.9	682.0	398.0	683.6	218.9	8078532.8	9201.7	9491.0	1316.7
	Standard Deviation	2.8	3.4	2.8	1.9	212.8	6.3	207.3	1.8	51390.2	28.2	130.8	8.2
	Coefficient of Variation	0.5	1.6	0.9	0.9	31.2	1.6	31.2	0.8	0.8	0.3	1.4	0.8
	Count Limit 3 sigma	0.01	0.05	0.03	0.03	0.94	0.05	0.94	0.02	0.02	0.01	0.04	0.02
Spin check 15/02/2003													
1		4245	13601	2989	13631	3500	11095	1631	8586	5829	9157	10719	11380
2		4211	13528	3025	14728	3911	11377	1568	8644	5921	9170	10737	11236
3		4211	13714	2989	14429	3924	11506	1549	8627	5886	9357	10970	11348
4		4239	13788	3010	13886	3577	11478	1533	8744	5918	9168	10657	11415
5		4225	13763	2987	13840	3604	11229	1554	8731	5884	9208	10906	11284
	Mean	4228.4	13678.8	3008.1	14145.4	3706.3	11396.6	1545.1	8868.2	5883.8	9213.8	10798.8	11333.0
	Standard Deviation	15.7	110.4	17.1	410.9	202.2	173.3	11.5	66.8	38.6	88.1	130.8	71.7
	Coefficient of Variation	0.4	0.8	0.6	2.9	5.5	1.5	0.7	0.8	0.7	1.0	1.2	0.6
	Count Limit 3 sigma	0.01	0.02	0.02	0.09	0.16	0.05	0.02	0.02	0.02	0.03	0.04	0.02
16-Feb-03													
6		4198	13731	2980	14079	3585	11138	1542	8539	5883	9110	10813	11305
7		4118	13568	2952	13909	3686	11041	1538	8618	5749	9044	10640	11150
8		4154	13364	2938	13620	3669	11189	1541	8558	5907	9008	10531	11100
9		4125	13320	2989	13658	3538	11159	1504	8524	5732	8890	10504	11045
10		4173	12951	2893	13585	3583	10957	1488	8428	5789	8949	10428	10890
	Mean	4153.1	13392.8	2952.8	13808.0	3567.8	11095.9	1522.3	8553.3	5785.8	9001.8	10543.3	11098.0
	Standard Deviation	32.8	283.0	40.3	213.6	20.5	95.8	25.0	83.5	63.1	82.1	85.5	151.8
	Coefficient of Variation	0.8	2.1	1.4	1.5	0.8	0.9	1.7	1.0	1.1	0.9	0.8	1.4
	Count Limit 3 sigma	0.02	0.06	0.04	0.05	0.02	0.03	0.05	0.03	0.03	0.03	0.02	0.04
Blank TE 15/02/2003													
1		0	0	0	0	19	6	22	1	16	2	8	0
2		0	0	0	0	18	5	21	1	15	2	8	0
3		0	0	0	0	17	5	20	2	15	2	7	0
4		0	0	0	0	16	5	18	1	16	2	7	0
5		0	0	0	0	18	5	18	1	16	2	8	0
	Mean	0.0	0.2	0.0	0.2	18.9	5.3	19.9	1.4	15.8	2.1	7.2	0.3
	Standard Deviation	0.0	0.0	0.0	0.0	1.5	0.2	1.5	0.1	0.3	0.1	0.7	0.0
	Coefficient of Variation	61.1	12.8	20.8	22.9	8.8	4.2	7.8	7.5	2.0	7.0	9.9	14.9
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
16-Feb-03													
6		0	0	0	0	15	6	17	1	16	2	6	0
7		0	0	0	0	14	5	18	1	16	2	6	0
8		0	0	0	0	14	5	17	1	16	2	6	0

APPENDIX EXPERIMENT M1

Run	Normalized Data	7Li	9Be	51V	52Cr	59Ni	60Co	60Ni	60Cu	68Zn	68Ga	75As	82Se	88Rb	88Sr	89Y	90Zr
8		8	0	82	271	44	23	110	25	28	20	14	5	13	25	1	90Zr
10		8	0	80	273	44	23	113	24	28	20	14	4	13	25	1	12
	Mean	7.6	0.5	83.6	274.4	43.9	22.8	111.7	25.3	28.4	20.4	14.3	4.8	12.7	25.1	1.0	13
	Standard Deviation	0.2	0.0	3.4	4.7	0.4	0.3	1.2	0.8	0.7	0.5	0.2	0.1	0.3	0.4	0.1	12.9
	Coefficient of Variation	2.5	8.7	4.1	1.7	0.9	1.3	1.1	3.1	2.5	2.3	1.7	1.7	2.1	1.8	5.5	3.5
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
SARM 1 15/02/2003																	
1		876	141	2800	4160	8008	129	180	881	3023	11700	758	15	142857	5478	96252	104528
2		861	140	2800	4113	63217	137	192	868	3051	11732	768	14	140280	5605	95103	103077
3		873	140	2481	4125	61858	142	185	868	3007	11390	768	15	138428	5378	85325	103207
4		865	140	2413	4088	63195	147	189	877	2998	11452	763	15	140351	5328	92819	102587
5		887	139	2378	4176	61938	151	187	880	3031	11680	764	15	141545	5334	94342	101882
	Mean	858.4	140.1	2530.7	4132.4	62558.9	141.2	188.5	880.3	3024.1	11580.8	763.1	14.8	140714.1	5404.0	94368.2	103047.8
	Standard Deviation	5.1	0.7	172.3	35.8	687.2	8.6	2.6	11.5	21.0	157.5	9.8	0.5	1677.8	82.2	1378.4	881.0
	Coefficient of Variation	0.7	0.5	8.8	0.9	1.1	6.1	1.4	1.3	0.7	1.4	1.3	3.2	1.2	1.5	1.5	1.0
	Count Limit 3 sigma	9.02	0.01	0.20	0.03	0.03	0.10	0.04	0.04	0.02	0.04	0.04	0.08	0.04	0.05	0.04	0.03
16-Feb-03																	
6		871	139	2383	4191	62352	156	187	901	3072	11852	778	14	138202	5393	92272	102872
7		872	141	2335	4113	62005	158	184	880	3010	12153	763	14	138187	5420	93853	101857
8		872	142	2347	4171	63173	163	184	884	3043	11858	762	15	142107	5444	96807	103817
9		871	140	2339	4136	62500	167	183	885	3045	11655	778	15	141184	5436	94801	104929
10		868	144	2335	4307	62280	187	182	880	3043	11823	768	15	138891	5452	82323	102587
	Mean	871.0	141.2	2342.0	4171.9	62483.9	162.2	184.1	890.0	3042.7	11748.5	777.1	14.8	140110.2	5428.9	93887.5	103212.6
	Standard Deviation	1.8	1.8	8.0	78.2	435.1	5.1	1.7	8.4	21.9	227.8	9.1	0.4	1564.4	23.3	1365.9	1189.2
	Coefficient of Variation	0.2	1.3	0.3	1.9	0.7	3.1	0.9	0.9	0.7	1.8	1.2	2.5	1.1	0.4	1.4	1.2
	Count Limit 3 sigma	0.01	0.04	0.01	0.08	0.02	0.09	0.03	0.03	0.02	0.08	0.03	0.07	0.03	0.01	0.04	0.03
	Average SARM 1	870	141	2438	4152	62561	152	186	885	3033	11690	773	15	140412	5416	94183	103130
	SARM Certified Value	12.00	7.75	2.00	12.00	154.89	0.36	8.00	12.00	50.00	27.00	18.30	0.01	325.00	10.00	143.00	300.00
	Counts per ppm	72	18	1218	348	404	421	23	74	61	433	40	1232	432	542	659	344
Concentrations in CRM's																	
Based on SARM 1																	
SARM 3 15/02/2003																	
Repeat		38	28	23	10	6481	2	13	13	348	54	8	<1	191	5172	26	11362
		38	28	23	10	6531	2	13	14	364	54	8	<1	191	5193	26	11375
SARM 48 15/02/2003																	
Repeat		14	1	50	411	10071	51	403	588	5316	12	879	<1	21	40	14	59
		14	1	50	405	8861	51	388	578	5226	11	865	<1	20	39	15	58
SARM 3 Cert Val																	
SARM 48 Cert Val		48.00	20.5	81	10	5953	2.44	2.20	13	385	54.00	1.92	0.01	180	4600	22	11000
				185	593		54	122	583	6200				18	28		95

APPENDIX EXPERIMENT M1

Run	Normalized Data	834B	804Mo	111Cd	126Sn	121Sb	126Te	138Ba	138La	140Ce	141Pr	146Nd	150Eu	157Gd	159Tb	163Dy	165Ho
9		15	3	0	5	1	0	881	0	0	0	0	1	0	0	0	0
10		15	3	0	6	1	0	882	0	0	0	0	0	0	0	0	0
	Mean	15.1	3.0	0.2	5.7	0.6	0.4	881.8	0.3	0.4	0.1	0.1	0.5	0.2	0.2	0.0	0.1
	Standard Deviation	0.4	0.1	0.0	0.2	0.0	0.1	8.3	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0
	Coefficient of Variation	2.9	3.4	17.0	3.8	4.7	12.7	1.0	11.2	4.8	14.8	18.0	14.3	15.7	28.7	34.2	23.6
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
SARM 1 15/02/2003																	
1		30012	441	24	1213	185	1	88329	108943	194833	27029	16782	233	3510	3718	6097	5485
2		30180	458	24	1431	188	1	78824	106804	180504	25568	18142	231	3483	3718	6130	5442
3		29899	437	24	1204	188	1	78517	106531	185588	26888	16241	228	3463	3802	6025	5383
4		29565	445	23	1196	185	1	80247	106221	191387	26380	16372	228	3448	3883	6165	5500
5		28855	442	25	1183	184	1	79453	107173	187208	26403	16185	230	3494	3887	6134	5389
	Mean	29822.5	444.2	23.9	1245.1	185.7	0.9	79778.0	107134.4	191686.2	26314.2	16336.8	229.9	3477.6	3881.8	6110.3	5440.9
	Standard Deviation	347.1	7.3	0.5	104.5	1.4	0.1	888.6	1070.3	2009.2	234.4	253.9	2.1	21.8	47.6	53.3	57.8
	Coefficient of Variation	1.2	1.7	2.2	8.4	0.7	10.6	1.1	1.0	1.0	0.9	1.6	0.9	0.6	1.3	0.9	1.1
	Count Limit 3 sigma	0.03	0.05	0.07	0.25	0.02	0.32	0.03	0.03	0.03	0.03	0.05	0.03	0.02	0.04	0.03	0.00
16-Feb-03																	
6		28943	441	23	1278	185	1	80420	107747	193935	26870	15969	225	3499	3632	8126	5426
7		29793	442	24	1201	185	1	77820	104333	188026	26217	15907	230	3512	3637	6040	5421
8		30159	447	24	1212	185	1	78162	105606	188710	26083	16176	229	3502	3694	6135	5397
9		29800	438	24	1186	184	1	78633	106823	189771	26202	16258	224	3488	3883	6135	5474
10		30142	441	24	1201	186	1	78804	106357	182158	26392	16188	227	3510	3636	8132	5445
	Mean	29859.2	441.9	23.9	1217.9	185.0	0.8	78541.9	105813.4	190400.0	26432.9	16107.7	227.2	3504.2	3888.4	6113.4	5432.6
	Standard Deviation	335.3	3.4	0.8	34.4	0.9	0.0	1852.0	1256.6	2329.5	384.8	148.0	2.5	8.3	30.0	41.0	28.8
	Coefficient of Variation	1.1	0.8	2.3	2.8	0.5	3.0	1.3	1.2	1.3	1.5	0.9	1.1	0.2	0.8	0.7	0.5
	Count Limit 3 sigma	0.03	0.02	0.07	0.08	0.01	0.08	0.04	0.04	0.04	0.04	0.03	0.03	0.01	0.02	0.02	0.02
Average SARM 1																	
	SARM1 Certified Value	28841	443	24	1232	185	1	79382	106524	191049	26534	16222	229	3481	3874	6112	5437
	Counts per ppm	53.00	2.84	0.11	3.30	1.18	0.01	120.00	109.00	195.00	18.50	72.00	0.35	14.00	3.00	17.00	3.60
		563	158	211	373	158	128	981	977	980	1381	225	663	248	1225	380	1510
Concentrations in CRM's																	
Based on SARM 1																	
SARM 3 15/02/2003																	
	Repeat	675	1	3	6	<1	<1	413	208	283	19	47	1	5	1	3	1
		653	1	3	6	<1	<1	414	208	281	18	48	1	6	1	3	1
SARM 46 15/02/2003																	
	Repeat	8	1	16	5	1808	<1	178	14	56	3	13	1	3	<1	2	<1
		8	1	16	5	1571	<1	172	14	56	3	13	1	3	<1	2	<1
SARM 3 Cert Val																	
	SARM 48 Cert Val	960	1.21	0.91	7.40	0.13	0.01	490	250	240	16	48	1.20	3.60	0.70	3.10	0.90
		28															

APPENDIX EXPERIMENT M1

Run	Normalized Data	166Er	169Tm	172Yb	175Lu	178Hf	181Ta	182W	205Tl	208Pb	209Bi	232Th	238U
8		0	0	0	0	14	5	16	1	15	2	6	0
10		0	0	0	0	14	5	17	1	15	2	6	0
	Mean	0.1	0.2	0.1	0.2	14.1	4.8	16.9	1.3	15.4	2.0	6.0	0.3
	Standard Deviation	0.0	0.0	0.0	0.0	0.8	0.2	0.6	0.1	0.3	0.1	0.2	0.0
	Coefficient of Variation	23.5	12.4	36.9	15.9	4.3	3.5	3.6	7.8	1.7	4.9	4.1	12.9
	Count Limit 3 sigma	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A
SARM 1 15/02/2003													
1		6015	2898	4425	2813	5625	7200	570	747	22056	305	58245	21264
2		6020	2884	4425	2858	5521	7221	565	748	22046	279	58957	21418
3		5925	2827	4422	2844	5328	7286	554	757	21512	283	58624	21307
4		5985	2854	4434	2859	5229	7183	563	771	22272	251	59784	21844
5		5916	2814	4398	2839	5116	7267	582	754	21238	256	59180	21439
	Mean	5874.1	2850.7	4420.9	2844.8	5363.7	7221.3	562.8	755.2	21824.7	270.7	59507.4	21450.6
	Standard Deviation	51.3	32.1	13.7	21.4	208.5	49.8	8.0	8.6	431.6	21.8	368.3	234.4
	Coefficient of Variation	0.9	1.1	0.3	0.8	3.9	0.7	1.1	1.3	2.0	8.1	0.8	1.1
	Count Limit 3 sigma	0.03	0.03	0.01	0.02	0.12	0.02	0.03	0.04	0.06	0.24	0.02	0.03
18-Feb-03													
6		5938	2829	4412	2880	5231	7228	570	754	21750	242	58618	21403
7		5892	2835	4440	2791	5222	7147	567	749	21548	253	57688	21193
8		5865	2805	4364	2829	5207	7175	584	757	21346	323	58343	21422
9		6035	2789	4334	2808	5178	7205	587	741	21804	390	58539	21247
10		6058	2862	4344	2829	5149	7324	582	751	22280	373	58857	21702
	Mean	5997.2	2824.1	4380.4	2827.3	5187.2	7216.0	588.2	750.8	21743.9	316.2	58574.5	21411.6
	Standard Deviation	58.0	27.9	48.3	33.6	33.7	67.7	3.0	6.1	352.8	87.8	687.5	203.6
	Coefficient of Variation	0.8	1.0	1.1	1.2	0.8	0.8	0.5	0.8	1.6	21.4	1.2	1.0
	Count Limit 3 sigma	0.03	0.03	0.03	0.04	0.02	0.03	0.02	0.02	0.05	0.84	0.04	0.03
Average SARM 1													
	SARM1 Certified Value	5886	2837	4401	2836	5280	7222	584	753	21784	293	58081	21431
	Courts per ppm	10.50	2.00	14.20	2.00	12.40	4.90	1.45	0.89	44.90	0.28	51.00	15.00
		570	1418	310	1418	426	1474	389	810	545	1067	1159	1428
Concentrations in CRM's													
Based on SARM 1													
SARM 3 15/02/2003													
	Repeat	2	<1	3	<1	224	13	5	<1	47	1	59	14
		2	<1	3	<1	225	12	5	<1	48	1	60	14
SARM 4B 15/02/2003													
	Repeat	1	<1	1	<1	1	<1	1	<1	14680	8	8	1
		1	<1	1	<1	2	<1	2	<1	14834	8	8	1
SARM 3 Cert Val													
	Er	Im	Yb	Lu	Hf	Ta	W	Pb	Th	U			
	2.80	3.00	0.40	231.00	25.20	0.53	43	0.47	68	14			

APPENDIX EXPERIMENT M1

Run	Normalized Data	7Li	9Be	51V	52Cr	55Mn	58Co	60Ni	65Cu	66Zn	68Ga	75As	82Se	85Rb	88Sr	89Y	90Zr
	Sample diluted 250x prior to analysis																
	Calculated																
	Detection Limit Data																
	Based on standards:-																
	Conc in ppb	7Li	9Be	51V	52Cr	55Mn	58Co	60Ni	65Cu	66Zn	68Ga	75As	82Se	85Rb	88Sr	89Y	90Zr
		10	24	13	28	6	9	30	15	8	6	13	55	8	8	6	28

APPENDIX EXPERIMENT M1

Run	Normalized Data	93Nb	88Mo	111Cd	120Sn	121Sb	126Te	138Ba	139La	140Ce	141Pr	148Nd	153Eu	157Gd	159Tb	163Dy	165Ho
	Sample diluted 250x prior to analysis																
	Calculated																
	Detection Limit Data																
	Based on standards:-																
	concs in ppb	93Nb	88Mo	111Cd	120Sn	121Sb	126Te	138Ba	139La	140Ce	141Pr	148Nd	153Eu	157Gd	159Tb	163Dy	165Ho
		8	9	9	6	6	18	8	7	5	7	8	5	8	7	4	5

APPENDIX EXPERIMENT M1

Run	Normalized Data	168Er	169Tm	172Yb	176Lu	178Hf	181Ta	182W	205Tl	208Pb	208Bi	232Th	238U
	Samples diluted 250x prior to analysis												
	Calculated												
	Detection Limit Data												
	Based on standards:												
	Concs in ppb	168Er 8	169Tm 8	172Yb 5	176Lu 5	178Hf 24	181Ta 39	182W 75	205Tl 8	208Pb 8	208Bi 9	232Th 5	238U 8

- 107 -

WE CLAIM:

1. Sample collection device comprising an inert collection matrix capable of adsorbing or absorbing a fluid sample, and a solid support, wherein the inert matrix is affixed to an area of the solid support.
- 5 2. A device according to claims 1, wherein the collection matrix is selected from the group consisting of aragonite, aluminum hydroxide, titania, glucose, Starch "A", Starch "B", glucodin, cellulose powder/granules, fibrous cellulose, hydroxy butyl methyl cellulose, vegetable flour or mixtures thereof
3. A device according to claims 2, wherein the vegetable flour is selected from the group consisting of rice, maize, wheat, soy, rye and corn flour, or mixtures thereof.
- 10 4. A device according to any one of the preceding claims, wherein the collection matrix is fibrous cellulose.
5. A device according to claim 4, wherein the fibrous cellulose matrix is modified by oxidation and/or acid hydrolysis.
- 15 6. A device according to any one of the preceding claims, further comprising, on or within the matrix, one or more pre-calibrated selected analytes as internal standard.
7. A device according to claim 6 wherein the pre-calibrated analytes are represented by or selected from the sets:
Li, Na, Mg, Al, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Cd, Sn,
20 Sb, Te, Ba, La, Ce, Eu, Dy, Yb, Hg, Tl, Pb, Bi, Th and U;
Li, B, Mg, Al, Si, P, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Sr, Y, Zr, Mo, Ag, Cd, Sn, Sb, Ba, La, Ce, Hf, Hg, Pb and U or
Li, Na, Mg, Al, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Cd, Sn, Sb, Te, Ba, La, Ce, Eu, Dy, Yb, Hg, Tl, Pb, Bi, Th and U.
- 25 8. A device according to any one of the preceding claims, further comprising a test sample.
9. A device according to claim 8, wherein the support comprises a bar-code incorporating information on the sample.
10. A device according to any one of the preceding claims, further comprising an integral lancing member, capable of piercing skin or tissue, to aid in the collection and application of a sample to the inert matrix.
- 30 11. A device according to claim 10, wherein the lancing member is mounted adjacent to, within or below the area of inert matrix.
12. A device according to claim 10 or claim 11, further comprising a guiding channel in the inert matrix, to guide the lance when the lance is disposed below the inert matrix area.
- 35

- 108 -

13. A device according to any one of the preceding claims, further comprising an integral or separate cover sheath, which covers the matrix.
14. A sample collection device having multi-layer construction wherein the collection matrix layer is sandwiched between two supporting layers, one of said supporting layers having an opening, which exposes an area of the collection matrix.
15. A device according to any one of the preceding claims, wherein the sample is a fluid sample selected from body fluids, oils and water.
16. A device according to claim 15, wherein the body fluid is selected from whole blood, urine and sweat.
17. Method of detecting simultaneously a plurality of elements in a fluid sample adsorbed onto or into an inert collection matrix, comprising:
- (i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample, and
 - (ii) detecting plurality of elements in the ionised portion of the sample by mass spectrometry.
18. Method of quantifying simultaneously a plurality of elements in a fluid sample adsorbed onto or into an inert collection matrix, comprising:
- (i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample;
 - (ii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;
 - (iii) measuring quantity of ionised portion of sample, and
 - (iv) determining quantity of the plurality of elements in the sample with reference to the quantity of ionised portion of the sample.
19. Method of quantifying simultaneously a plurality of elements in a fluid sample adsorbed onto or into an inert collection matrix having an internal standard applied thereto, comprising:
- (i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample and a portion of said internal standard;
 - (ii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;
 - (iii) measuring quantity of ionised internal standard in the ionised portion of the sample by mass spectrometry, and
 - (iv) determining quantity of the plurality of elements in the sample with reference to quantity of ionised internal standard.

- 109 -

20. Method of quantifying simultaneously a plurality of elements in a fluid sample adsorbed onto an inert collection matrix, comprising:

(i) introducing into the fluid sample a known quantity of a measurable internal standard

5 (ii) exposing the sample to high energy radiation capable of ionising at least a portion of the sample and the internal standard;

(iii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;

10 (iv) measuring quantity of ionised internal standard in the ionised portion of the sample by mass spectrometry, and

(v) determining quantity of the plurality of elements in the sample with reference to quantity of ionised internal standard.

21. Method of quantifying simultaneously a plurality of elements in a fluid sample adsorbed/absorbed onto or into an inert collection matrix comprising:

15 (i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample;

(ii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;

20 (iii) exposing a matrix-matched Certified Reference Material (CRM) to high energy radiation capable of ionising at least a portion of the CRM;

(iv) measuring quantity of ionised CRM in the ionised portion of the sample by mass spectrometry, and

(v) determining quantity of the plurality of elements in the sample with reference to the CRM.

25 22. Method of quantifying simultaneously a plurality of elements in a fluid sample supported on an impermeable substrate, comprising:

(i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample;

30 (ii) measuring quantity of a plurality of elements in the ionised portion of the sample by mass spectrometry;

(iii) exposing a matrix-matched Certified Reference Material (CRM) to high energy radiation capable of ionising at least a portion of the CRM;

(iv) measuring quantity of ionised CRM in the ionised portion of the sample by mass spectrometry, and

- 110 -

(v) determining quantity of the plurality of elements in the sample with reference to the CRM.

23. A method according to claim 19 or claim 20, wherein the internal standard is selected from the group consisting of Li, Na, Mg, Al, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Cd, Sn, Sb, Te, Ba, La, Ce, Eu, Dy, Yb, Hg, Tl, Pb, Bi, Th, and U.

24. A method according to claim 19 or claim 20, wherein the internal standard is selected from the sets:

Li, Na, Mg, Al, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Cd, Sn, Sb, Te, Ba, La, Ce, Eu, Dy, Yb, Hg, Tl, Pb, Bi, Th and U;

Li, B, Mg, Al, Si, P, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Sr, Y, Zr, Mo, Ag, Cd, Sn, Sb, Ba, La, Ce, Hf, Hg, Pb and U or

Li, Na, Mg, Al, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Rb, Sr, Mo, Cd, Sn, Sb, Te, Ba, La, Ce, Eu, Dy, Yb, Hg, Tl, Pb, Bi, Th and U.

25. A method according to claim 21 or claim 22, wherein the CRM is selected from the group consisting of SARM 1, 3 and 46, and SY-2.

26. A method according to any one of claims 17 to 24, wherein the inert collection matrix is part of a sample collection device according to any one of claim 1 to 14.

27. A method according to any one of claims 17 to 26, wherein the fluid sample is selected from body fluids, oils and water.

28. A method according to claim 27, wherein the body fluid is selected from whole blood, urine and sweat.

29. A method according to claim 28, wherein the sample is whole blood and sample size is about 50 μ l to about 100 μ l.

30. A method according to claim 28, wherein the sample size is about 50 μ l or less.

31. A method according to any one of claims 17 to 30, wherein the high energy radiation is UV laser radiation.

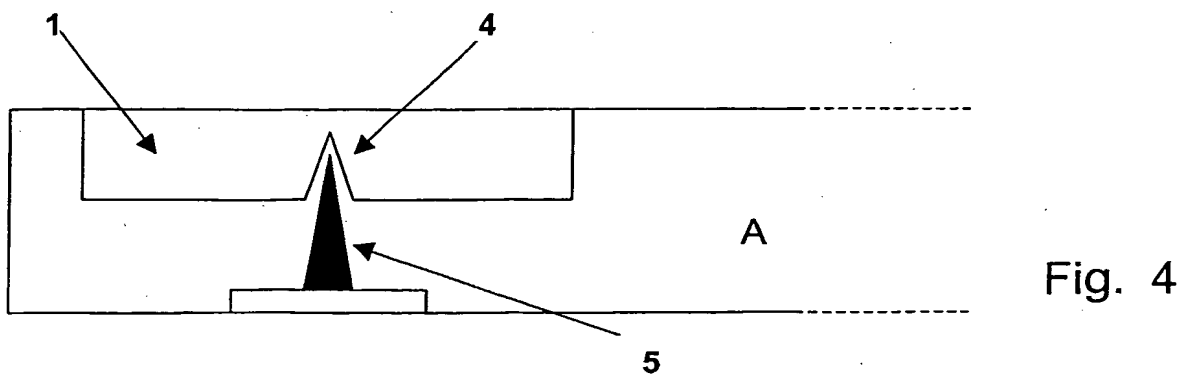
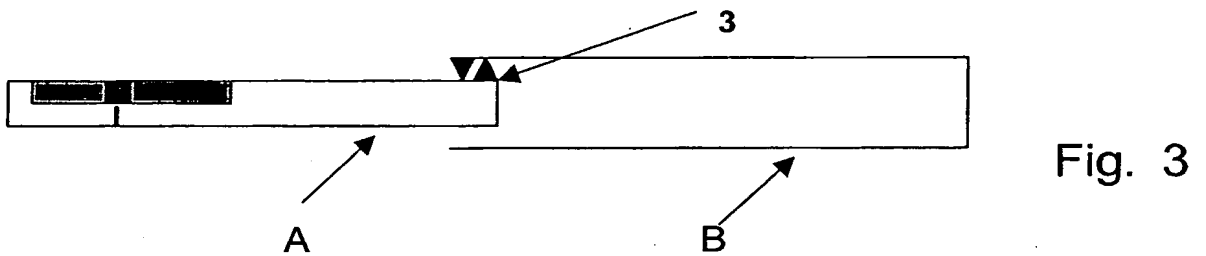
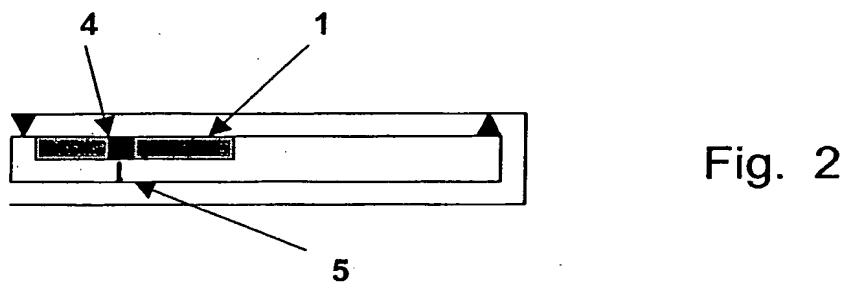
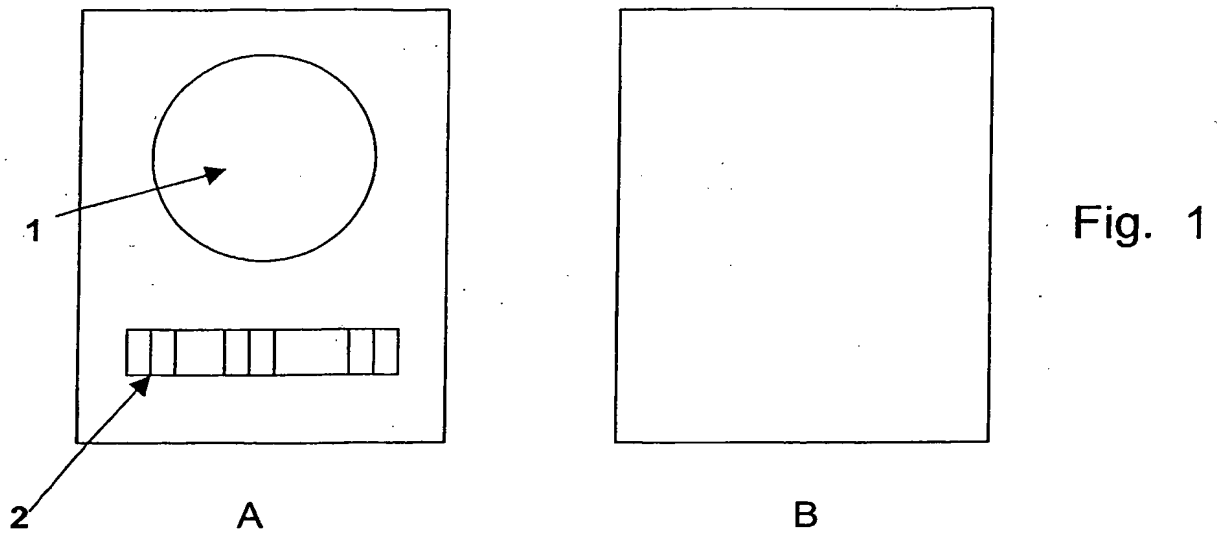
32. A method according to claim 31, wherein the laser radiation is a component of Inductively Coupled Plasma-Mass Spectrometer (ICP-MS).

33. A method according to claim 32, wherein the mass spectrometer is selected from quadrupole and Time-of-Flight (TOF).

34. A method according to any one of claims 17 to 33, wherein the sample is exposed to radiation for a period of from about 10 seconds to about 120 seconds.

- 111 -

35. A method according to any one of claims 17 to 34, wherein the elements to be detected and/or quantified are selected from dietary trace elements, toxic elements and markers of pollution or wear and tear.
36. A method according to any one of claims 17 to 34, wherein the matrix or the support comprise one or more wells or indentations to accommodate the fluid sample.
37. A method of collecting a fluid sample for mass spectrometry analysis of multiple element content comprising the application of the sample to an inert matrix having a low background element content, wherein the matrix is selected from the group consisting of aragonite, aluminum hydroxide, titania, glucose, Starch "A", Starch "B", glucodin, cellulose powder/granules, fibrous cellulose, hydroxy butyl methyl cellulose, vegetable flour or mixtures thereof.



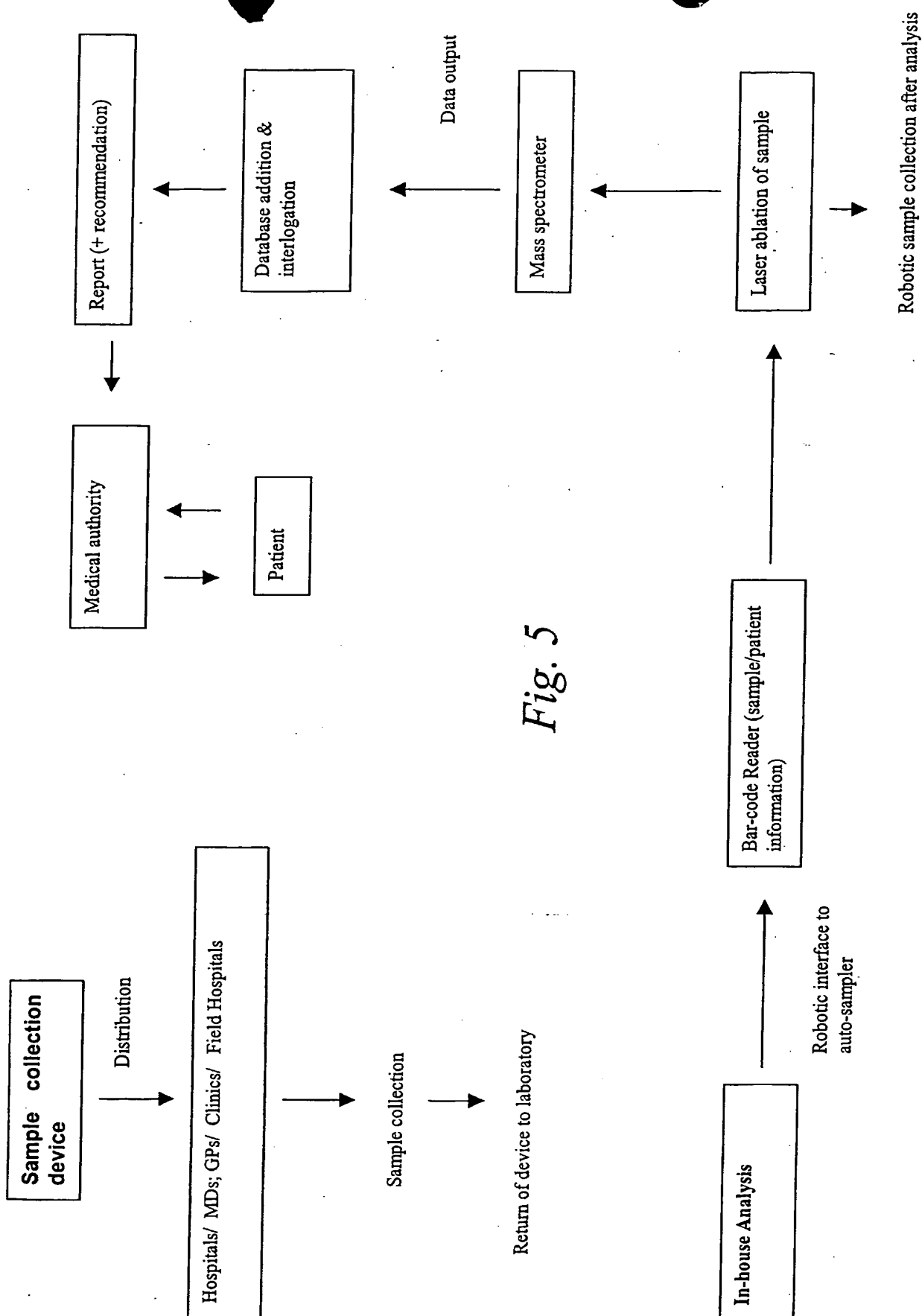


Fig. 5

Rec'd PSTA 14 OCT 2004

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU03/00450

A. CLASSIFICATION OF SUBJECT MATTER

Int. Cl. ⁷: G01N 1/10, 30/72, 33/487

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
DWPI: (blood or sample) and (analyte or matrix) and (lance or pierce or needle or sharp) and layer and (hole or opening or aperture) or (mass spectr+ and (many or lots or plurality) with sample) and (reference or standard))

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 179 005 A (PHILLIPS et al) 12 January 1993 See figs.	1,2,37
X	DE 201 18 772 U1 (8SENS BIOGNOSTIC AG) 28 March 2002 See figs.	1,2,4,10,13-16,37
X	US 6 124 012 A (JONES JR et al) 26 September 2000 See abstract.	1,13-16
X	EP 852 336 A (LIFESCAN, INC) 8 July 1998 See claims.	1,2,13-16,37

☒ Further documents are listed in the continuation of Box C

☒ See patent family annex

* Special categories of cited documents: document defining the general state of the art which is not considered to be of particular relevance	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"E" earlier application or patent but published on or after the international filing date	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"O" document referring to an oral disclosure, use, exhibition or other means	"&" document member of the same patent family
"P" document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search
18 June 2003

Date of mailing of the international search report
26 JUN 2003

Name and mailing address of the ISA/AU
AUSTRALIAN PATENT OFFICE
PO BOX 200, WODEN ACT 2606, AUSTRALIA
E-mail address: pct@ipaustalia.gov.au
Facsimile No. (02) 6285 3929

Authorized officer

SUSAN T. PRING
Telephone No : (02) 6283 2210

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU03/00450

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 345 781 B (BEHRINGER MANNHEIM CORP) 13 December 1989. See figs.	1,2,13-16,37
X	EP 715 337 B (HITACHI LTD) 14 March 2001 See claims.	17,18,26-36
X	WO 94/28418 A (BAYLOR COLLEGE OF MEDICINE) 8 December 1994 See abstract.	17,18,26-36
X	EP 738 000 B (BRUKER DALTONIK GMBH) 16 February 2000 See claims.	17,18,26-36
X	WO 96/03768 A (VESTEC CORP) 8 February 1996 See abstract.	17,18,26-36
X	WO 01/94910 A (BASF AG) 13 December 2001 See abstract	17-36
X	US 2001/013579 A (ADRIEN JR et al) 16 August 2001 See abstract.	17-36

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU03/00450

Box I Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos :
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos :
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos :
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a)

Box II Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

See supplemental sheet.

1. ☒ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/AU03/00450

Supplemental Box

(To be used when the space in any of Boxes I to VIII is not sufficient)

Continuation of Box No:

The international application does not comply with the requirements of unity of invention because it does not relate to one invention or a group of inventions so linked as to form a single general inventive concept. In coming to this conclusion the International Searching Authority has found that there are two inventions:

1. Claims 1-16 are directed to a sample collection device attached to a support. Claim 37 is a method claim for collecting a sample by using the sample collecting device as stated above. It is considered that a sample collecting device attached to a support or a method of using the aforesaid comprises a first "special technical feature". Their classification would nominally be G01N 1/10. The dependent claims of claim 1 add additional features that from the description appear to be mere embodiments.

2. Claims 17-36 are directed to a method of detecting simultaneously a plurality of elements in a fluid sample adsorbed/absorbed onto or into an inert collection matrix or supported on an impermeable substrate comprising:

(i) exposing the sample to high energy radiation capable of ionising at least a portion of the sample, and

(ii) detecting plurality of elements in the ionised portion of the sample by mass spectrometry.

It is considered that exposing the sample to high energy radiation capable of ionising at least a portion of the sample prior to the step of detecting a plurality of elements in the ionised portion of the sample by mass spectrometry comprises a second "special technical feature". Their classification would nominally be G01N 30/72, 33/487.

Consequently the common features do not constitute "a special technical feature" within the meaning of PCT Rule 13.2, second sentence, since it makes no contribution over the prior art. Since there exists no other common feature which can be considered as a special technical feature within the meaning of PCT Rule 13.2, second sentence, no technical relationship within the meaning of PCT Rule 13 between the different inventions can be seen. Consequently it appears that a posteriori, the claims do not satisfy the requirement of unity of invention.

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/AU03/00450

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report	Patent Family Member	
US 5 179 005	NONE	
DE 201 18 772	NONE	
US 6 124 012	NONE	
EP 852 336	AU 45307/97	JP 10-191995
EP 345 781	JP 2-059648	
EP 715 337	JP 8-145950	
WO 94/28418	EP 700 521	JP 2000-131285
EP 738 000	NONE	
WO 96/03768	US 5 498 545	EP 771 470
WO 01/94910	AU 69058/01	
US 2001/013579	US 6 541 768	
END OF ANNEX		

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ **BLACK BORDERS**
- ☐ **IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- ☐ **FADED TEXT OR DRAWING**
- ☐ **BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- ☐ **SKEWED/SLANTED IMAGES**
- ☐ **COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- ☐ **GRAY SCALE DOCUMENTS**
- ☐ **LINES OR MARKS ON ORIGINAL DOCUMENT**
- ☐ **REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- ☐ **OTHER: _____**

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.